Universidade de Aveiro Departamento de Ambiente e Ordenamento
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FÉLIX RICARDO CHARVET MALDONADO

DESEMPENHO DOS PELLETS DE BIOMASSA EM SISTEMAS DE COMBUSTÃO DOMÉSTICA

PERFORMANCE OF BIOMASS PELLETS IN DOMESTIC COMBUSTION SYSTEMS

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Dissertação apresentada à Universidade de Aveiro para cumprimento dos requisitos necessários à obtenção do grau de Mestre em Sistemas Energéticos Sustentáveis, realizada sob a orientação científica do Professor Doutor Luís António da Cruz Tarelho, Professor Associado do Departamento de Ambiente e Ordenamento da Universidade de Aveiro

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agradecimentos

Me gustaría expresar mis agradecimientos a todas las personas que de una u otra manera fueron parte importante durante esta etapa de mi vida.
A mis padres Ricardo y Liz, y a mi hermana Estefanía por su infinito amor y apoyo incondicional.
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Gracias totales.

palavras-chave Combustão, biomassa, fogão, desempenho, pellets.

resumo

Nos últimos anos, o uso dos pellets de biomassa como fonte de energia alternativa apresentou um crescimento constante. O processo mais difundido para a conversão termoquímica de biomassa é a combustão. Pellets certificados e comerciais, e pellets de biomassa não comerciais e não certificados, foram utilizados neste trabalho. Oito tipos diferentes de pellets de biomassa foram testados num fogão a pellets com uma potência nominal de 9,5 kW_{th}, tendo sido caracterizadas as condições de operação do processo de combustão. O fogão utilizado da marca Solzaima modelo Alpes, fabricado em Portugal, é uma tecnologia para aquecimento doméstico. Para monitorizar e realizar a aquisição das variáveis operatórias experimentais durante as experiências de combustão foi desenvolvido um software. O trabalho focalizou-se em caracterizar os parâmetros operatórios do processo de combustão no fogão, tais como a temperatura de combustão em cinco locais distribuídos ao longo da câmara de combustão e da chaminé, o consumo de combustível, o caudal de ar de combustão, as necessidades estequiométricas de ar de combustão, o excesso de ar, e a composição dos gases de combustão. Foi determinado o balanço de carbono para o processo de combustão dos pellets de biomassa testados, com o objetivo de determinar a eficiência de conversão de carbono. Observou-se que as características físico-químicas dos pellets e o sistema de alimentação do fogão que não proporciona uma alimentação verdadeiramente contínua e uniforme, influenciam o desempenho do processo de combustão, nomeadamente em termos de parâmetros de processo e composição dos gases de exaustão. No caso de algumas experiências de combustão de pellets, a emissão de gases poluentes ultrapassa os limites estabelecidos pela legislação portuguesa para instalações de combustão. Além disso, observou-se como o processo de paragem do fogão a pellets criou condições de combustão que promovem a emissão de poluentes relacionados à combustão incompleta, tais como CO, C_2H_6 e CH_4. As características dos pellets de biomassa como o alto teor de cinzas ou de umidade, foram identificadas como desfavoráveis.

keywords Combustion, biomass, stove, performance, pellets, pollutants.

abstract

In recent years the use of biomass pellets as an alternative energy source has experienced steady growth. The most widespread process for thermochemical conversion of biomass is combustion. Standardized commercial pellets and non-commercial non-standardized biomass pellets were used in this work. Eight types of biomass pellets were tested during combustion experiments in a pellet stove with a nominal power of 9.5 kWth, and the operating conditions were characterized. The stove used was a Solzaima model Alpes made in Portugal, a technology used for domestic heating. In order to monitor and acquire the experimental data during the combustion process, an interface software was developed. The work focused on characterizing the operating parameters such as the combustion temperature at five locations distributed along the combustion chamber and the chimney, the biomass fuel consumption, the flow rate of combustion air, the stoichiometric air needs, the excess fuel-air ratio, and the flue gas composition. Carbon balance was evaluated for every biomass pellets tested to determine the carbon conversion efficiency. It was observed that the physical and chemical characteristic of the pellets and the feeding system of the stove which does not provide a true continuous and uniform feeding have a direct influence over the operating parameters and therefore, in the composition of the exhaust gases. In some cases, gaseous pollutant emissions overpassed the limits set by Portuguese regulations. Also, it was observed that the shut-down procedure of the stove created combustion conditions that promote the emission of pollutants related to incomplete combustions such CO, C2H6, and CH4. Characteristics of pellets such as the high ash or moisture content were identified as unfavorable.

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Nomenclature

A	Transversal area of the chimney	[m^2]
b	Carbon balance	[-]
$\overline{Cy}$	Mean concertation of (y =CH_4, C_2H_6, SO_2, NO) in combustion flue gas	[mg · Nm^3]
EFy	Emission factor (y = CH_4, C_2H_6, SO_2, NO)	[mg y · kg^{-1}_{db}]
$\dot{m}_{air}$	Primary air flow rate	[Nm^3 · h^{-1}]
$\dot{m}_{c,CO}$	Carbon mass flow content in CO	[kg $_C$ · h^{-1}]
$\dot{m}_{c,CO_2}$	Carbon mass flow content in CO_2	[kg $_C$ · h^{-1}]
$\dot{m}_f$	Fuel flow	[kg $_{fuel\ db}$ · h^{-1}]
$\dot{m}_{c,in}$	Carbon mass flow in	[kg $_C$ · h^{-1}]
$\dot{m}_{c,out}$	Carbon mass flow out	[kg $_C$ · h^{-1}]
$\dot{m}_{c,TOC}$	Carbon mass flow content in TOC	[kg $_C$ · h^{-1}]
M_j	Molar mass (j = C, O, N, S, H,)	[kg · mol^{-1}]
M_{H_2O}	Water molar mass	[kg mol^{-1}]
M_{N_2}	Nitrogen molar mass	[kg $_N$ · mol^{-1}]
M_{O_2}	Oxygen molar mass	[kg $_{O2}$· mol^{-1}]
t	Time	[h]
TOC	Total organic carbon	[mg to_C · Nm^{-3}]
u	Combustion flue gas velocity	[m · s^{-1}]
$\overline{\dot{V}_{fg}}$	Combustion flue gases mean flow	[m^3· h^{-1}]
W_c	Carbon mass fraction per kg fuel	[kg $_C$ · kg^{-1} $_{fuel\ db}$]
Wj	Mass fraction (j = C, O, N, S, H,) per kg of fuel	[kg $_j$ · kg^{-1} $_{fuel\ db}$]
w_s	Stoichiometric combustion oxygen	[kg O_2 · kg^{-1} $_{fuel\ db}$]
W_{of}	Oxygen mass fraction per kg of fuel	[kg O_2 · kg^{-1} $_{fuel\ db}$]
w_{SA}	Stoichiometric combustion air	[Nm^3 $_{air}$ · kg^{-1} $_{fuel\ db}$]
W_{V_A}	Moisture ratio in the combustion air	$\left[kg_{H_2O} \cdot kg_{air}^{-1}\right]$
W_w	Water mass fraction per kg of fuel	[kg $_C$ · $kg^{-1}$$_{fuel\ as}$]
Y_{sj}	Stoichiometric oxygen consumption for element j	$\left[kmol_{O_2} \cdot kmol_j^{-1}\right]$
z	Excess air	[%]
λ	Excess air ratio	[-]

Index

as	As received
db	Dry basis
HC	Hydro Carbons
TOC	Total organic carbon

1. Introduction

The process of social, economic and technological transformation since the mid- seventeenth century, revolutionized the industry, trade between countries, changed the way in which humans perform daily activities, and contributed to the development of science and technology. The technological transformation led to the mechanization of processes in the industry introducing mass-production of goods. This transformation also changed the means of transportation and improved the quality of life of people thanks to electricity, internal combustion engines or air conditioning systems.

As a consequence, this new way of living of humanity has brought the exponential demand of energy resources. Fossil fuels have been and remain as the more demanded energy resource. Worldwide, around 85% of the energy used in the four primary energy consuming sectors, (i.e., industrial, commercial, residential, transport) comes from fossil sources such as oil, coal or natural gas [1]. Meanwhile, in Europe, 77% of the average European energy requirements are covered with fossils fuels [2].

The activities carried out by humanity in these four sectors have a direct link with the environmental pollution. For example, the emission of combustion gases as a result of burning fossil fuels for the electricity generation and the pollutants emission generated in the transportation sector are the main responsible for global warming. Nowadays, the current patterns of energy use have a significant impact on the environment, and their effects are discernible[2] [3].

Humanity has begun to take actions to mitigate the effects of pollution. The challenge is to find a way that allows the development of humanity in a way that meets the needs of the population in the present without compromising the ability of future generations to meet their own needs.

To achieve this goal, renewable energy sources such as bioenergy has entered into the global energy scenario to replace traditional fossil sources. Bioenergy is the energy derived from biological sources. It emerges as a solution to minimize the dependence from fossil sources. Biomass is bioenergy produced in green plants, either terrestrial or aquatic, which through photosynthesis converts sunlight into plant material and stores the energy of the sun in the chemical bonds of carbohydrates [4].

Biomass is an exciting solution to achieve sustainable development because it is a form of non-intermittent energy, which can be supplied in high quantities, with relatively low costs, and promoting local economic structures generating additional income for farmers[5]. Biomass can be used to produce various bioproducts such as ethanol, fuel pellets, biodiesel or syngas[6].

The use of biomass contributes to the reduction of emissions gases responsible for the greenhouse effect. The use of biomass is considered carbon neutral since the CO_2 emissions released in the processes of transformation of biomass into thermal energy, is compensated by the absorption of that CO_2 in the regrowth of the plants [7].

For of all these favorable characteristics, the use of forest biomass for thermal energy production has experienced an increase on a the global scale [8]. However, the operational

combustion parameters, and the physical and chemical characteristic of the biomass used as fuel, influence the emission of atmospheric pollutants[9]. Therefore, it is essential to characterize the operational parameters related with the combustion of biomass in different combustion technologies. This work emerges as an opportunity to experiment, test and characterize the operation conditions of the combustion of biomass pellets with a technology used for domestic heating.

1.1 Objectives.

The objective of this work was to characterize the combustion performance of eight types of biomass pellets by using a domestic technology: A stove for domestic heating. The following activities were developed:

- Review of the literature on the production of biomass pellets, their characteristics, and energy conversion by combustion.
- Development of interface software for control and data acquisition.
- Combustion experiments using biomass pellets in a stove for domestic heating and characterization of the operating conditions.
- Treatment and analysis of information collected.

2. Literature Review

2.1 Biomass as bioenergy

Biomass is a solid fuel constituted by organic matter from animals and plants such as wood from forests, crops, seaweed, material left over from agricultural and forestry processes as well as the biodegradable fraction of industrial and urban waste. Biomass is considered a renewable energy source because it has a supply that is not limited. It is always posible to grow trees and crops, and waste will always exist [10] [11] [12].

Photosynthesis is the process in wich biomass transforms carbon dioxide from the atmosphere, water, and sunlight into organic matter. Organic matter includes molecules of cellulose, hemicellulose, lignin, lipids, HC, sugars and other components. The energy of the sun is stored in the form of chemical energy in the bonds of these molecules. Cellulose and hemicellulose are the molecules with a greater significant value because these chains provide to the biomass specific characteristics [1] [13].

The energy of the sun stored in the chemical bonds of carbohydrates is called bioenergy, and it can be recovered by various process of transformation. Compared to other renewable technologies, such as solar or wind, biomass has fewer problems with energy storage; in some way, biomass is stored energy [1][14].

Biomass has the potential to provide sustainable energy in the future while helping countries achieve their greenhouse gas reduction targets [15][16].

2.2 Woody biomass

Woody Biomass denotes all biomass from conifers or deciduous; angiosperms or gymnosperms, and included stems, branches, foliage, bark, chips, lumps, granules, briquettes, sawdust, sawmill and others [11] [17]. Around the 64% of the energy obtained from biomass derives from wood and wood waste [18] which, gives biomass from wood a significative relevance. The most predominant chemical elements in the composition of woody biomass are, carbon, hydrogen, oxygen, nitrogen, and sulfur. In a dry ash free basis analysis, carbon shows as the most predominant element in biomass from wood [11] (**Table 1**).The biomass analyzed in this study comes from woody species derived from forest sources available in Portugal, Italy and Spain, such as pine, eucalyptus , acacia, and poplar.

Table 1. Chemical composition of some varieties of woody biomass in dry base analysis without ash.[11][17][19][20][21]

Variety Wood Biomass	**C**	**O**	**H**	**N**	**S**
	%wt	%wt	%wt	%wt	%wt
Eucalyptus Bark	48.7	45.3	5.7	0.3	0.05
Pine Bark	53.8	39.9	5.9	0.3	0.07
Mean Value Woody Biomass	52.1	41.2	6.2	0.4	0.08

Besides the chemical composition of woody biomass, other properties inherent to it determine the most optimal choice for the thermochemical conversion. Such properties also have an influence in the operating parameters prompting the emissions of pollutants or rising the costs of processing and operation of the conversion technology. The properties are related to the following aspects: (i) intrinsic and extrinsic moisture content, (ii) calorific value, (iii) fixed and volatile carbon proportions, (iv) ash content (v) content of alkali metals, (vi) cellulose/lignin ratio [13]. For thermochemical conversion processes of dry woody biomass, the first five properties have a special interest [22].

2.3 Woody biomass pre-treatment methods

Woody Biomass pre-treatment methods refers to all processes to convert harvested wood or residual wood into standardize biofuel with similar physical and chemical properties and characteristics in order to guarantee a certain quality, better than the one obtained from raw wood biomass (without treatment).

The characteristics required or desired in a woody biomass are; (i) homogenization of the fuel, which allows the use of an automatic biofuel feeding system, thus reducing the investment, maintenance, personnel, and operation costs of the conversion unit. (ii) energy density increase, thus reducing the storage cost and the handling of the biofuel. (iii) the reduction of the impurities contained in the biofuel (for example, stones, earth, sand, metal parts, glass, plastics)[22] [23]. This study analyzed two pre-treatments methods; pelleting and torrefaction.

2.3.1 Torrefaction of woody Biomass

Torrefaction is a method that improves the quality and properties of woody biomass. It turns the woody biomass more suitable to be used as biofuel in a thermochemical

conversion process. The torrefaction process consists in a slow heating of the woody biomass within inert atmosphere (atmosphere without the presence of O_2) up to a maximum temperature of 575 K [24]. This lower range of temperature compared with other woody biomass pre-treatment process like pyrolysis, allows biofuel to retain most of its energy content (around 90%). In addition, woody biomass loses an approximately 70% of its original dry weight. The final results obtained with this method are; a solid uniform biofuel with lower moisture and higher energy content than raw wood [15] [25].

Torrefaction brings to the biofuel improved properties; (i) increased hydrophobicity, which makes the fuel water-repellent and more resistant to biodegradability allowing the storage of biofuel for longer periods without degradation of quality, (ii) improvement in the grindability, therefore, milling the biofuel in smaller particles needs less energy compared to milling of raw biomass [26], (iii) the calorific value, increases due to a decrease in hydrogen and oxygen content [15] [26].

2.3.2 Pelleting method

Woody biomass such as sawdust, wood shavings, wood chips, wood trunks, forest residues, straw biomass and stems are the raw materials for the manufacture of wood pellets [27] [28]. Raw woody biomass is characterized for its low energy density ($MJ \cdot m^{-3}$) and low mass density ($kg.m^{-3}$) [29]. The purpose of the pelleting method is to convert raw woody biomass into a solid biofuel keeping a certain quality, a constant size, low moisture content, high energy density, and similar physical dimensions. Lower emission of pollutants is an advantage of the combustion of pelletized biomass over the combustion of raw biomass for the production of thermal energy [29] [30].

2.3.2.1 Description of the pelleting method

The pelleting method of wood biomass involves three main operations; (i) reduction in size by crushing, (ii) raw wood biomass drying (iii), densification (pelleting) [31] [32]. The entire process is described below (**Figure 2.1)**.

2.3.2.2 Size reduction

The first step is to clean the raw material. This procedure removes heavy contaminants such as rocks, metals and other foreign materials. Subsequently, the cleaned raw material goes through a crushing process to reduce its size. The objective of the crushing is also to homogenize the size of the raw material [29].

2.3.2.3 Drying

Moisture has a considerable influence on the final result and on the amount of energy required to manufacture wood pellets. Raw material needs to dry until the content of moisture is less than 10%wt [31]. Raw materials with a water content of more than 15% are more difficult to pelletize [29], and also demand an extra amount of energy in the dry process. The energy required for drying in some cases is generated by the same raw material used to feed

the pelleting section, in this way a reduction of the economic and energy costs, and operating under the values of sustainable development is achieved [27]. The drying process is not necessary if the raw material comes from the furniture industry, where the moisture content is already low [29].

2.3.2.4 Densification

Immediately after the drying process, the dry material passes through another size reduction process conducted at a hammer mill. The objective here is to obtain raw material particles with a suitable size, between 3.2 mm to 6.4 mm [29]. Subsequent particles are pressed by the action of rollers, followed by extrusion through a perforated matrix with circular holes. In general, this process occurs at elevated temperatures, because of friction and pressure caused by the force of the rollers in the matrix. The temperature reached has relevance in the process because, when the wood lignin (woody biomass component) is heated, it acquires certain plasticity facilitating the aggregation of the particles for the formation of pellets [33]. This phase of the process accounts for most of the electricity consumption in pellet manufacturing and the primary source of maintenance costs [29].

The pellets are cooled immediately after the pelletizing process to adjust the additive bond and stabilize its shape. The result is cylinders with typical dimensions of Ø 6-10 mm that can be used in stoves or boilers with automatic loading due to their good fluidity, uniform water content, and size [22]. The pellets can be stored in silos or packaged according to their final purpose. For domestic or small-scale use, 15 kg bags are normally used. For industrial or large-scale, 700-1000 kg bags are used [29] [30].

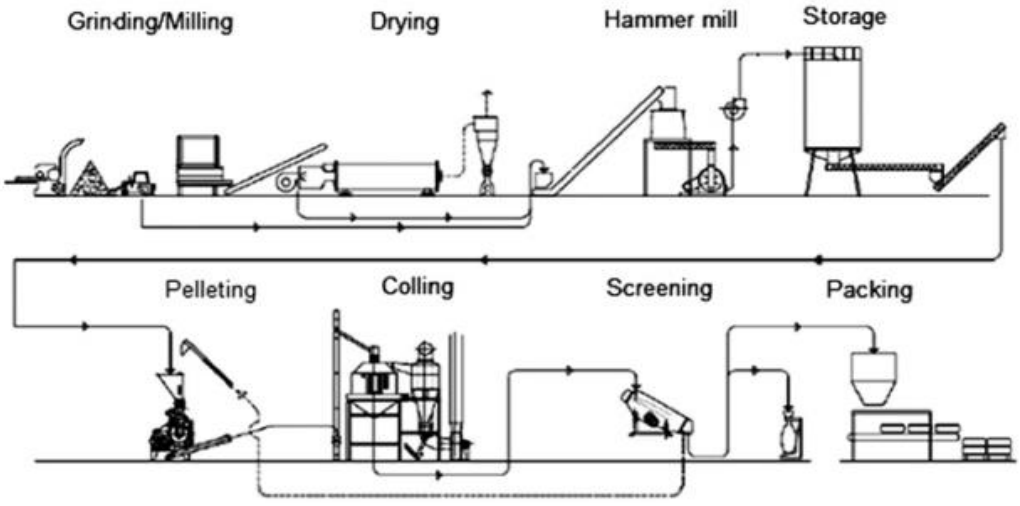

Figure 2.1 Schematic of the pelleting process [29].

2.3.3 Pellet's regulation / standardization

Standardized pellets are a biofuel that must meet specific quality standards for the commercialization and use within the European territory. In Europe, there are five known standards: NF Biofuels Solides (France), DINplus (Germany), O NORM (Austria), SS 18 71 20 (Switzerland) and EN PLUS (Europe). This standardization certifies the entire pelleting

process, including logistics. The European standard EN PLUS has three degrees: ENplus-A1 and ENplus-A2 (residential/institutional use) and EN-B (industrial use). Raw materials need to be carefully selected to accomplish the standards[34][35]. **Table 2** shows the properties that pellets must fulfill according to the European standard EN PLUS.

Table 2. Some of the properties of pellets by the EN PLUS standardization normative [35].

	Diameter	Length	Moisture Content	Ash Content	Net Calorific Value	Sulfur Content
	mm	mm	w-% [2]	w-% [2]	MJ/Kg [1]	w-% [2]
ENplus-A1	6 or 8	3.15 ≤ L ≤ 40	≤ 10	≤0.7	16.5 ≤ Q ≤ 19	≤0.03
ENplus-A2	6 or 8	3.15 ≤ L ≤ 40	≤ 10	≤1.5	16.3 ≤ Q ≤ 19	≤0.03
EN-B	6 or 8	3.15 ≤ L ≤ 40	≤ 10	≤3.0	16.0 ≤ Q ≤19	≤0.04

1)As received 2) Dry Basis

2.3.4 Factors that influence the pelleting process

The pressure required for pelleting is a highly demanding energy factor, hence its importance to control it. The pelletizing pressure is a variable that is in function of the biomass chemical and physical properties, the moisture content and the particle size desired. Pelleting pressure decreased with increasing moisture content of the raw material. [36].

Other factor that influence the pelleting process is the ash content in the biomass. The presence of ash in biomass increase the cost of production and affects the quality of the final product, generating operations problems during the combustion. The ash content is directly related to the amount of bark contained in the raw material Pellets made from harvest residues (branches and tops) have a high bark content and as a consequence high ash content [34].

2.3.5 Advantages of biomass pellets

Pelleting gives to the raw woody biomass some properties, which are beneficial for its uses as biofuel. The energy density of biomass pellets are four times higher than the energy density in raw wood chips [27] [32] (**Table 3).** The benefit of a higher the energy density is the reduction of volume required for transport and for storage of the pellets compared with raw biomass. That reduces the transport costs of pellets, making more attractive the possibility for distributing pellets at greater distances from the manufacturing point; therefore, pellets could be used in large urban areas and large public buildings at a regional or international scale [27] [34] [28].The standardization of its composition, humidity, and size allows automatic feeding in domestic and industrial technologies reducing operating costs [36].

Table 3. Properties of wood pellets and raw wood chips [27].

	Calorific Power		Water	Apparent Density	Ash Content
	GJ / t	kWh / m^3	%	kg / m^3	%wt
Wood Pellets	17.0	3080	8	650	0.5
Wood Chips	13.4	750	25	200	1

The storage of pellets under the control of relative humidity, temperature, and ventilation, allows preservation of more than five years without significant degradation [37]. In an uncontrolled environment, its durability is around five months [38].Finally, concerning the perception of users for their use as biofuel, wood pellets are considered highly acceptable because people have used wood as fuel for a long time [37]. For the indicated advantages, wood pellets, as a source thermal energy, is considered one of the most promising alternative energy source [30].

2.3.5.1 Pellets moisture content

The moisture content of the biomass is one of the most unfavorable characteristics of the fuel. While the oxidation reactions in the combustion process are exothermic, the evaporation of the water is endothermic. Therefore, it affects the self-sufficiency of the combustion process [39]. To maintain the self-sufficiency of the combustion process the moisture content of the biomass pellets cannot be higher than 65%wt (wet basis) [40]. Above this point, the energy released is insufficient to compensate the evaporation and heating of the product[4].

2.3.5.2 Pellets heating value

The heating value (HV) is an expression of the energy content in the biofuel, usually measured regarding the energy content per unit mass, for solid fuels ($MJ \cdot kg^{-1}$). The HV of a fuel can be expressed in two forms, the higher heating value (HHV) and the lower heating value (LHV). The LHV is the total amount of heat evolved in the complete combustion of fuel without accounting for the amount corresponding to the latent heat of condensation. The water formed will be in gaseous phase and thus the energy required to vaporize the water, is not released as heat. The HHV is the total amount of heat evolved in the complete combustion of the fuel when the water vapor originating in the combustion is condensed. Thus, heat released in this phase change is counted [13] [17]. The lower heating value is essential because determines the use of any material as fuel. The heating value of a wood fuel decreases with increasing the moisture content of the wood [41].

2.4 Conversion processes

Chemical energy stored in the biomass can be converted into other forms of energy or products through various processes using different technologies [13] [42]. The products are; (i) thermal energy / heat, (ii) biofuels for transportation, (iii) chemical feedstock [4] [43]. There

are three main processes to make the conversion; (i) thermochemical conversion, (ii) biochemical / biological conversion, (ii) mechanical conversion by stratification technology [4] [39] [44].

The thermochemical conversion process includes four options for biomass conversion into energy; (i) combustion, (ii) pyrolysis, (iii) gasification, (iv) liquefaction [4] [42] [39]. In the combustion process, the energy stored in the biomass is released as thermal energy/heat. In the pyrolysis, liquefaction or gasification processes the chemical energy stored is transformed into a solid product as charcoal or liquid products such as fuel or gaseous product such as synthetic gas (Syngas) [43]. The form in which energy is required for it final use determines the process, and the conversion technology chosen [4] [39].

The biochemical/biological conversion process involves two options, (i) digestion, which could be summarized as the production of biogas composed mainly of a mixture of methane (CH_4) and carbon dioxide (CO), (ii) the fermentation process used to produce ethanol for use as a biofuel [4][43]. Thermochemical processes have an advantage over biochemical/biological. The reaction time required for a thermochemical process are a few seconds or minutes, whereas biochemical/biological processes take several days, weeks or even longer, and not all the biomass is suitable for biochemical conversion [39] [45] [46]. **Figure 2.2** resumes the biomass conversion process into energy.

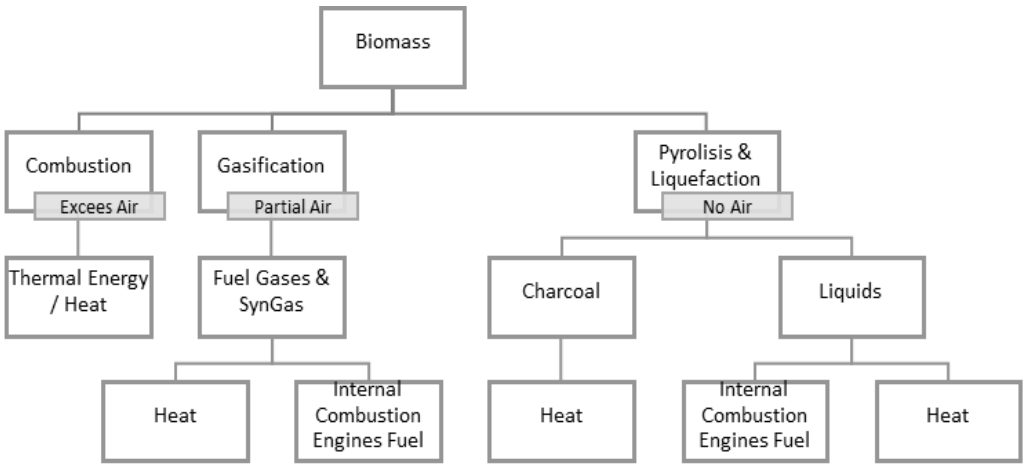

Figure 2.2. Diagram of thermochemical conversion processes [4] [47] [48].

2.4.1 Combustion

The combustion process is the most used process for the conversion of biomass into thermal energy. Around the 97% of the bioenergy produced in the world, comes from the combustion of biomass. The process of conversion by combustion is considered reliable, proven, low cost, relatively well understood, and has a wide range of commercially technology available [39] [41] [49]

The process of combustion consists of a series of thermochemical reactions that lead to an exothermic oxidation produced at a relatively elevated temperature. The process oxidizes

the carbon (C) and the hydrogen (H) content in the biomass into CO_2, and H_2O [49] [50] .During the combustion process, the biomass particles undergo five stages: (i) heating and drying, (ii) devolatilization and volatile combustion, (iii) gasification, swelling and primary fragmentation, (iv) char combustion with secondary fragmentation, (v) oxidation of gas phase components (e.g., volatiles) [51] [50].The factors that affected the duration of each of these phases depends on the physical and chemical properties of the fuel, the temperature and the combustion operating parameters [51].The second stage (Devolatilization and volatile combustion) is responsible for the contribution of more than 70% of the total heat generation of the entire process. Devolatilization and volatile combustion is carried out above the fuel bed and is generally evident by the presence of yellow flames. The combustion of biomass produces hot gases at temperatures in a range between 800°C to1000 °C [4] [39] [52].

2.4.1.1 Stoichiometric air ratio

To guarantee an efficient conversion of the solid fuel, enough oxygen must be provided to the combustion process. Inside the combustion chamber, the air-fuel mixture is far from the ideal conditions and thus, some parts inside the combustion chamber can be oxygen starved. Therefore, excess oxygen is always provided to the combustion chamber [50]. One of the main operational parameters of the combustion process is the ratio between the amount of air provided to the combustion system and the stoichiometric amount of combustion air for that specific fuel. This relation is express in terms of an alternative parameter, the coefficient lambda (λ). Insufficient combustion air leads to incomplete combustion and consequently the formation of many products of incomplete combustion (pollutants). Excess air also cools the system reducing the amount of recoverable energy. The stoichiometric air requirements depend on the chemical and physical characteristics of the fuel [41]. Combustion operation parameters of the combustion technologies influence the emission of pollutants released to the atmosphere. For example, the CO emission, in a small domestic technologies present higher emission of CO, than the industrial technology.

2.4.2 Gasification

Gasification is a thermochemical conversion in which a solid or liquid material, with a carbon base, is converted into a combustible gas. The process involves the use of gasifying agents such as; steam, water, air, carbon dioxide, pure oxygen, or a mixture of these gases [53].

The process is carried out at elevated temperatures to optimize gas production. The gas produced is composed by a mixture of H, CO, CO_2, CH_4 and other hydrocarbons, as well as nitrogen, sulfur and other compounds [54]. The fuel submitted to gasification conversion process could be coal, oil, or biomass. In biomass gasification, the first step of the process is the thermochemical breakdown of cellulose, hemicelluloses and lignin compounds with the production of carbon and volatile substances [55]. The composition of the gas produced depends on factors such as; process conditions, fuel properties, gasification agents, temperatures of the conversion equipment. The purpose of gasification is the production of a gas with a particular heating value that can be used as fuel in a combustion engine to produce power[48].

2.4.3 Pyrolysis

Pyrolysis is a process that can be described as the direct thermal decomposition of wood biomass carried out in an inert atmosphere [23]. It is carried out at temperatures between 400°C to 800°C [56]. Typically, the pyrolytic gas is captured and burned in a supplementary combustion chamber, to obtain the heat required for the process [57].

The pyrolysis products are gas, liquid and solid carbon. The rate of pyrolysis influences the final products. The slow-rate pyrolysis produces solid carbon (biochar)as the primary product, while fast-rate pyrolysis produces liquid carbon (bio-oil) as the primary product and biochar as a sub-product. The proportions of the gas, liquid and solid carbon obtained depend on the method of analysis, the characteristics of the biomass and the reaction parameters. [23] [58] Although pyrolysis can be considered an efficient pre-treatment for wood biomass [58], at the same time it is particularly challenging because it develops many physical and chemical transformations and produces various species of products [9].

2.4.4 Liquefaction

Liquefaction is the conversion of biomass into a stable liquid hydrocarbon using low temperatures and high hydrogen pressures[4].

2.5 Combustion technologies

Combustion can be done through the use of a wide range of technologies available in the market. Nowadays stoves, ovens, boilers, steam turbines, turbo generators are available to convert the chemical energy stored in biomass into heat, mechanical energy or electricity[41]. The main advantage of the devices used nowadays is the energy conversion efficiency. For example, a modern wood stove is 10% or 20% more efficient than an old-fashioned wood stove. Hence the interest is its widespread use based on the action plan of several countries for the efficient use of energy[59][22][41]. The next section is divided into biomass combustion technologies for domestic use and biomass combustion technologies for industrial use.

2.5.1 Conversion technologies for domestic use

For hundreds of years, the process of burning fire-wood has been used as a source to provide heat for domestic proposes and, during those hundreds of years, the technology evolved. Traditional devices such as simple chimneys or stoves, have been gradually replaced by highly sophisticated electronically controlled combustion systems, designed for the combustion of wood pellets, wood log, wood chips [22].The most common types of domestic combustion technologies are summarized below.

2.5.1.1 Wood stoves

A wood stove is an autonomous device used to heat the space within which it is located, without the use of ducts [22]. They are simple devices without a sophisticated control system and low efficiency. The combustion rate of wood stoves is regulated by controlling the supply of primary combustion air. The fuel (wood-logs) is supplied manually. The operator controls both processes. Wood stoves transfer heat by radiation and convection. In many

stoves, a viewing window is provided in the front door, in order to gives the operator a much better opportunity to adjust the stove for optimal combustion [59].

2.5.1.2 Wood pellet stoves

Nowadays, there are stoves specially designed for the combustion of biomass pellets for residential heating. The nominal power range is between 2.5kW$_{th}$ to 25kW$_{th}$. These stoves have quite extensive electronic control systems, so they need to be connected to electricity for operation, unlike a traditional fire-wood stove. The electronic control provides advantages; for example, the combustion air is controlled through the action of an electric fan. The benefit of combustion air control is reflected in low emissions of CO and C_xH_x (hydrocarbons). For fuel storage, pellet stoves usually have a small deposit. The transport of the deposit to the combustion chamber is carried out through an electronically controlled feeding system. The fuel supply speed can usually be adjusted and varies according to the heat demand. The combustion air is extracted through slots in the inner wall of the housing through an air extractor, which reduces the pressure in the combustion chamber. The combustion gases flow towards the chimney passing before by a heat exchanger. The "cold "air in the room enters the stove near the bottom of the unit and leaves heated on top of the stove. Pellet combustion stoves can achieve efficiencies of more than 80% percent [22] [60]. The required maintenance is minimal, and the devices can remain operational for more than a week without the manual removal of ashes [22].

2.5.1.3 Pellet burners for central heating systems

Pellet burners for centralized heating use are based on the same operating principle as pellet stoves and have the same control systems. The difference lies in two aspects: (i)the heat is captured as hot water not warm air, (ii) the nominal power, these pellet burners built for a nominal thermal power of up to 25kW$_{th}$ [22].

2.5.2 Operating conditions in domestic technologies

Regardless of the technology used to biomass combustion, polluting gases are emitted into the atmosphere (**Figure 2.3**). These pollutant gases have a negative effect on air quality at various spatial scales. The effects on human health and the environment of these gases are of public domain [30] [37].

Domestic combustion technology and industrial combustion technology have intrinsic operating conditions. For example, the technologies for domestic use operated with low temperatures in the combustion chamber, and restrictions in the mixture of fuel and oxygen [61] [62]. These particular operating conditions promote the emission of a high quantity of some pollutants, mainly CO. To minimize this operational disadvantage and control the emission of CO, the domestic combustion technology operates with an excess of air relative to the stoichiometric superior to 200% [61] [63]. In one hand operation with excess air guarantees a good mixture of inlet air and fuel, but on the other one, excess air drops the combustion temperature, as a consequence the efficiency of the process in the domestics combustion technology is reduced [22] [64].Efficiency domestic technologies (Pellets stoves) are between 68% to 89% [62] [65].

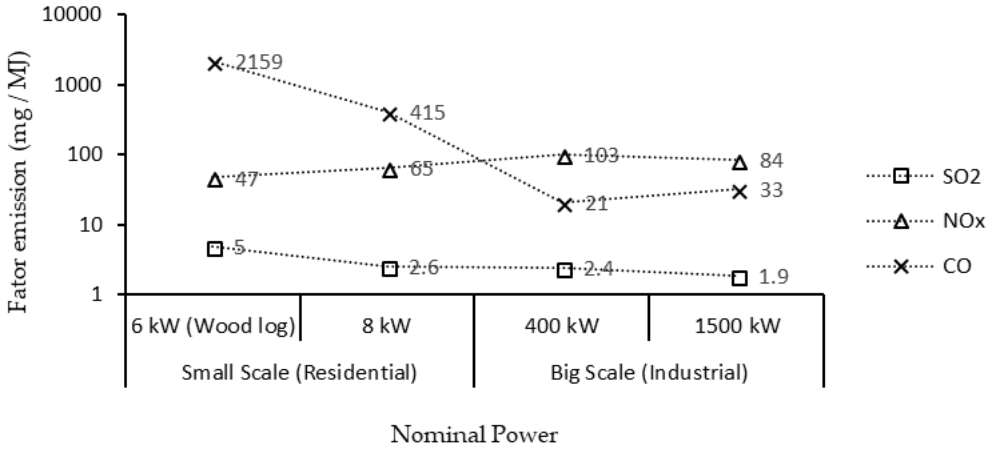

Figure 2.3 Comparation between the emission factors of SO_2, NOx, CO with the residential and industrial technologies based in the power output [66][67].

2.5.3 Conversion technologies for industrial use

The combustion technologies commonly exceed the $100kW_{th}$ of nominal power. These devices are equipped with systems that fully control operation. It is possible to distinguish between three combustion technologies for industrial use; (i) fixed bed combustion; (ii) fluidized bed combustion; (iii) pulverized fuel combustion[22][50].

2.5.3.1 Fixed-bed combustion

Fixed-bed combustion systems include fixed grates, moving grates, traveling grates, rotating grates and vibrating grates. In fixed bed combustion, the primary air passes through a fixed-bed, where the combustion process takes place (dry, gasification and the combustion of the fuel). The produced combustible gases are burned after the addition of secondary air, and it generally occurs in a combustion zone separated from the fixed-bed. This is called staged combustion and is a characteristic of fixed- bed. The staged combustion separate gasification and oxidation zones and avoids back-mixing of the secondary air. Grid furnaces are suitable for biomass fuels with high moisture content, variable particle sizes and a high ash content [50] [51].

2.5.3.2 Fluidized-bed combustion

The combustion systems in fluidized bed combustion (FBC) have been applied since 1960 in the combustion of municipal and industrial waste. The particularity of the FBC reactors is that the fuel is burned in a self-mixing suspension of air bubbles, fuel particles, and bed material. The bed material represents 90-98% of the fluidized bed. The BCF's provide suitable conditions for complete combustion with low demand for excess air, due to the high rate of transfer of heat into the mixture. The combustion within the BCF reactors allows the use of diverse types of biofuel mixtures from sources such as woody, agricultural, urban waste. Depending on the fluidization velocity, it is possible to distinguish two types of BCF; the bubbling fluidized bed (BFB) and the circulating fluidized bed (CFB). For fuels with

particles up to 40 mm in size, CFB's are used, while for particles less than 80 mm, BFB's are used. Fluidized bed combustion systems require a long time for reach operating temperature (approximately 8 to 15 hours) for which gas or oil burners are used [50] [68] [69].

2.6 Environmental impacts

Polluting gases emitted during combustion of biomass have an adverse effect on air quality at various spatial scales. The effects of the gases on human health and the environment are of public domain [30] [37]. The gases are described below.

2.6.1 CO_2

The CO_2 emissions in combustion processes are justified by the fact that more than 95% of the carbon present in wood pellets is converted into CO_2 during the combustion process [37][30]. Nevertheless, CO_2 emissions from biomass combustion are considered CO_2-neutral. This is the main environmental benefit of biomass combustion[57] [70] [71].

2.6.2 NO_x

Nitrogen oxides are originated mainly from anthropogenic sources. Combustion processes are the primary generator of NOx. Nitrogen oxides disrupt the normal vegetation growing. In the case of human being's, NOx affects the respiratory system and prevent oxygen to be transported to red cells. At a mesoscale NOx contributes to the formation of photochemical smog [72]. Most of the time NOx emissions from biomass combustion applications are a result of complete oxidation of nitrogen content in the fuel. For temperatures in the combustion chamber below to 1300 °C as in pellets stoves, the fuel NOx mechanism is the main responsible from the NO and NO_2 formation. Typically, N in the fuel is converted into >90% of NO and >10 % of NO_2. [37][61]. The NO concentration in the combustion flue gas increases with increasing nitrogen content in the fuel, excess air ratio and combustion temperature, up to a point where all fuel nitrogen intermediates have been converted to either NOx, N_2O or N_2 [22] [73].

2.6.3 SO_x

The combustion of fuels from wood sources emits sulfur into the atmosphere in the form of SO_2 and SO_3. Immediately after the combustion process 99% of the sulfur contained in the fuel is released in the form of SO_2 and the rest in the form of SO_3 [68]. The combustion of wood pellets has a lower SO_2 emission compared to other biofuels [37] [74] [75].

2.6.4 HCl

Hydrogen chloride is a toxic compound with that causes corrosion in material and have a negative effect on the human respiratory system. The presence of HCl in combustion flue gases, is directly related to the chlorine content in the biomass pellets. The emission of HCl may be more evident from biomass pellets containing higher amounts of chlorine, such us grass and straw. [22][75].

2.7 Emission from incomplete combustion

Emissions caused by incomplete combustion are mainly a result of either; i) insufficient mixing of combustion air and fuel in the combustion chamber, which produces local fuel-rich combustion zones, ii) lack of available oxygen, iii) too low combustion temperatures, iv) too short residence times. The presence of CH_4, CO, NH_3, C_2H_6 in combustion flue gases are indicators of incomplete combustion [22] [50] [75].

2.7.1 CH_4 and C_2H_6

Methane and ethane are organic compounds that are emitted during the incomplete combustion of biomass due to the carbon content present in the fuel. The presence of methane and ethane in the combustion flue gas are indicators of incomplete combustion[37]. Improvements in the efficiency of the conversion technology, a correct control of the combustion air supplied, and elevated temperatures in the combustion chamber, lead to a reduction of methane and ethane emissions [75].

2.7.2 CO

Carbon monoxide emission sources can be biogenic, for example uncontrolled forest fires and anthropogenic as emission from engines in the transportation sector due to incomplete combustion of fossil fuels. [73]. The emission of CO depends on the composition of the fuel, the excess of air, and the combustion temperature. The conversion of the C in the fuel into CO_2 occur through numerous stages, the last of these stages is the oxidation of CO into CO_2 ,if enough oxygen is available. The temperature at the combustion chamber regulates the frequency of conversion; therefore the concentration of CO in the combustion flue gas is higher when the combustion temperature usually decreases below 800 °C [75][37]. A higher excess air relative to the stoichiometric will ensure enough oxygen available nevertheless, will reduce the combustion temperature; thus, a mid-point needs to be found.

2.7.3 NH_3

Small amounts of NH_3 may be emitted as a result of an incomplete conversion of NH_3, formed from pyrolysis/gasification to oxidized nitrogen-containing components. This occurs with low a temperature in the combustion chamber [22] [75].

2.8 Pellets life cycle

When considering the pellets life cycle (logging, production, distribution, and combustion) of wood pellets for the generation of thermal energy, results presented that although, pellets are not a neutral carbon energy source, their use has a positive impact on air quality and in the reduction of the greenhouse gases emission. Some studies reported a reduction of the greenhouse gases in a range between 82% to 92% for electricity generation with wood pellet, compared with fossils fuels [30] [70] [76]. During the pellets life cycle, the activity that generates the most significant environmental impact is the pellets making process, followed by pellet combustion by the final user, and finally the biomass transport [77]. The pellets production process is the most energy-intensive activity; consider an energy

balance, the energy used in this activity is around 17% of the energy contained in the wood pellets delivered to the customers [78].

Life cycle assessments have been used in calculating greenhouse gases emission impacts of pellets production chain. Results showed that the production of one ton of pellets with round wood logs using a motor-manual felling, delimbing with a chainsaw, timber yarding with a tractor along the forest track, and transportation of the logs up to a distance of 35km to the pellets factory emitted about 83 kg of CO_2eq [79] [80]. Life cycle assessments showed that better benefits in the use of biomass pellets are obtained when the supplying distance is less than 600 km [81]

Compared the used of biomass pellets in heating systems against the ones based on diesel, or natural gas, the biomass system benefits from reduced impacts on the greenhouse gases. However, the biomass system performs worse on the local impact such as particulate matter formation, human toxicity, photochemical ozone formation, and land use [77].

Although, the CO_2 emitted throughout the pellet life cycle will be reabsorbed during the process of re-growth of vegetation/ trees to produce the new biomass pellets. The re-growth process is longer than the pellet life cycle. This period of re-growth should be considered in the development of policies based on the use of biomass as an energy source [30] [70] [82] [83].

3. Experimental work

The experiments of pellets combustion were conducted by using a domestic technology; a stove for domestic heating, located in the Environment and Planning Department at Universidade de Aveiro.

3.1 Pellet stove for domestic application

The combustion experiments were carried out using a top-feed pellet stove with a nominal output of 9.5 kW_{th}, manufactured in Portugal by Solzaima S.A, model *Alpes* used for domestic heating. The stove is designed for used biomass pellets as fuel with a diameter of 6 mm and a length in the range of 10 to 30mm; others dimensions could affect the stove's performance [84]. The stove has an internal pellet storage tank with 20 kg capacity. A screw feeder supplies the pelletized fuel into the combustion chamber through a feeding port (**Figure 3.1**).

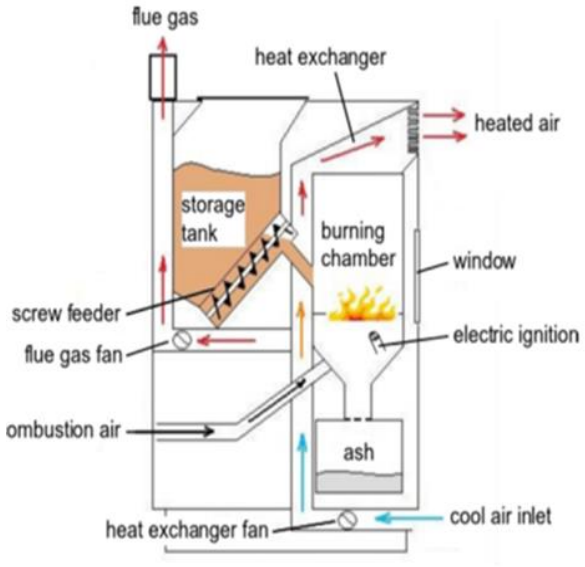

Figure 3.1. Operating principle of a pellet stove [85].

The pellet stove was putted on a scale with a 20 g resolution allowing the biomass fuel consumption to be monitored. The mass flow of fuel in the stove was calculated for each experiment by dividing the total amount of burned fuel by the total time of the combustion test. Primary air is supplied through holes in the bottom of the grate, and secondary air enter through holes located on the top of the combustion chamber. Primary air is driven by an electric fan located downstream of the combustion chamber. The primary intake air flow rate was measured continuously during the combustion using a mass flow meter (Kurz S.A model Adam).

Figure 3.2. Experimental facility. The stove mounted on the scale to measure the stove mass variation during a combustion experiment.

The stove has two operation modes; automatic, and manual. The manual mode allows the modification of two parameters, *Fire* and *Fan*. Fire is the level of power, and it can be modified in five levels. The level one is for minimum machine power up to mode five the maximum power. The levels power modifying feeding fuel rate to the combustion chamber. The *fan* parameter is the speed of the heat exchanger fan and has five levels of variation. The mode one is for minimum speed, and mode five is the maximum speed [84].

Figure 3.3 Control panel of the pellet stove. Manual mode allows the variation of five levels of fuel feed rate, and five levels of the heat exchanger fan velocity [84].

The oxygen concentration in flue gas (O_2) was measured using a zirconia oxygen sensor (OXY-FLEX, from SST Sensing Ltd). The temperature was measured continuously using K-type thermo-couples in 5 locations along the combustion chamber and the chimney. The **figure 3.4** shows the location of the three thermo-couples situated inside the combustion

chamber. The volumetric flow rate of combustion flue gases was determined, based on the monitoring (every 10 minutes) the combustion flue gas velocity at the top of the chimney with a Pitot tube connected to a differential pressure transducer. The stove is equipped with a vertical chimney with an internal diameter of 80mm and 4160 mm tall. Equation 1 was used to determinate the volumetric flow rate of combustion flue gases

$$\dot{V}_{fg} = u \times A \qquad \text{(Eq.1)}$$

Figure 3.4. Location of the thermo-couples T1, T2, T3 in the combustion chamber.

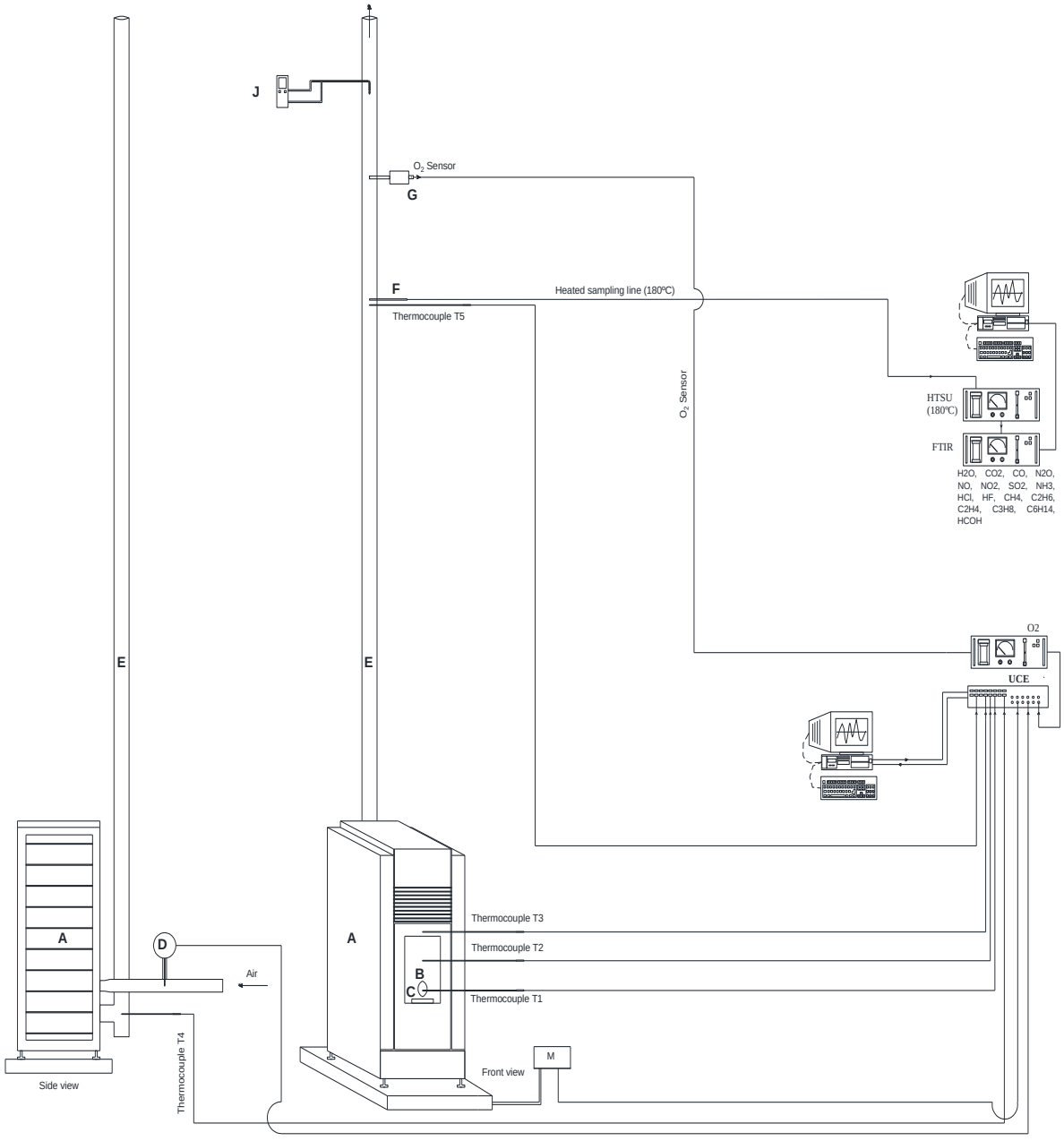

Figure 3.5. Layout of the experimental facility. A- Stove, B - Combustion chamber, C - Grate of the stove, D - Air flow meter, E - Exhaust duct (Chimney), F – Combustion flue gas sampling and analysis system, G –O_2 Sensor, J - Pitot tube, M - Scale.

The combustion flue gas was sampled at the exit of the chimney and conduced in a heated sampling line (180°C) to the gas analyzer (Gasmet, CX4000). This equipment has a multi-component measurement capability, which enables the real-time monitoring of Water (H_2O), Carbon dioxide (CO_2), Carbon oxide (CO), Nitrogen oxides (NO_x), Methane (CH_4), Ethane (C_2H_6), Hydrogen chloride (HCl), Sulphur oxide (SO_2), Formaldehyde (HCHO) and

Total organic carbons (TOC). The concentration of the exhaust gases was recorded as wet gas and then corrected to a dry basis based on the H_2O concentration measure. The flue gas composition data was acquired every 20 seconds.

3.1.1 Monitoring and control system

To monitor in real time the operating variables during the combustion experiments, an interface platform was developed for data monitoring and acquisition. Web Access software from Advantech S.A company in conjunction with two ADAM-4019+ universal analog input modules and one, ADAM-4561 addressable converter, were used for this purpose.

Eight analog inputs of 32 bits were used for monitoring and control the operation parameters of the stove. Five analogs inputs were used for temperature measurement by using k-type thermocouples located in five various locations**,** see **Figure 3.4** and **3.5.** One analog signal in a range between 0-1V for measure the primary air intake airflow rate, one analog signal in a range between 0-20mA was used for the acquisition data from the scale, and one analog signal in a range between 0-1V from for measure the O_2 concentration in the flue gases. All the analogs signals were converted into digital signals through the Adam 4561 addressable converter module and sent through a universal serial bus cable to a DELL computer using a Pentium dual-core of 2,25 GHz 2GB of RAM. Calibration curves were built for O_2, intake airflow meter and scale using references parameters. **Figures 3.6**, **3.7**, and **3.8** represent calibration curves and calibration equations.

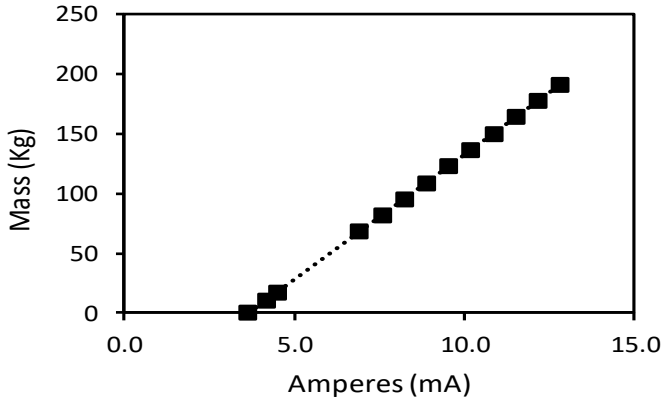

Figure 3.6. Calibration curve of the scale ($y = 20.82x - 75.963$; $R^2 = 0.99$).

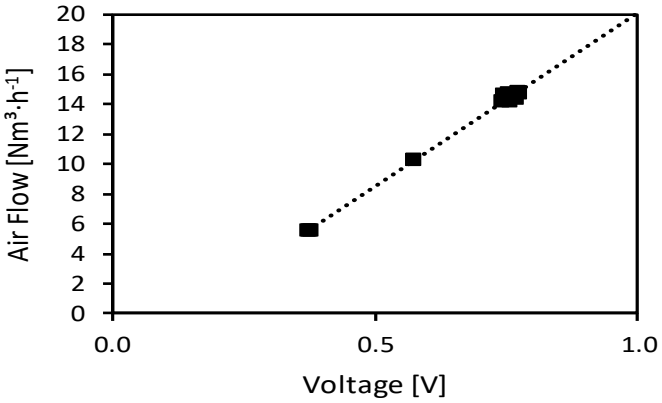

Figure 3.7. Calibration curve of the primary air intake flow sensor ($y = 23.31x - 3.146$; $R^2 = 0.99$).

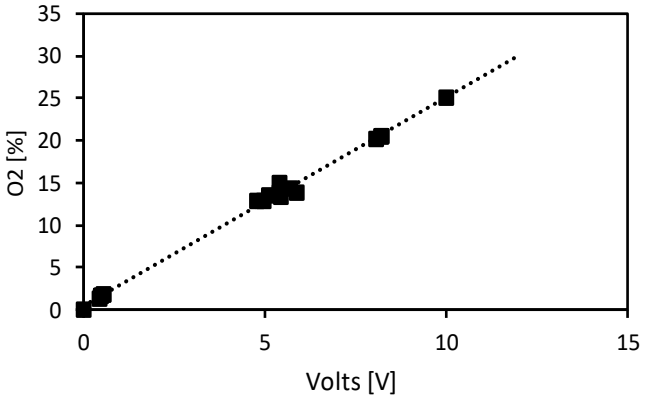

Figure 3.8. O_2 sensor calibration curve ($y = 2.463x + 0.439$; $R^2 = 0.99$).

WebAccess S.A software allowed the development of a graphic interface with charts and diagrams. A layout of the stove drew in AutoCAD was imported in order to have a graphic reference of the location of the 8 signals (sensors). Three gauges were designed to monitor the O_2 concentration in the flue gases, and also to control variation of mass in the scale, and visualize the primary air intake airflow rate (**Figure 3.9)**. A chart (**Figure 3.10**) was integrated into the interface to have the temperature profile at the five locations of the thermocouples in real time. The data acquired from all eight inputs was recorded synchronized in a single Excel file. Time of data acquisition could be modified according the operators pre-requisites. For all the combustion experiments in the stove, data was acquired every 20 seconds. The layout of the experimental facility showing all the devices to control and monitoring the combustion experiments is show in **Figure 3.5**.

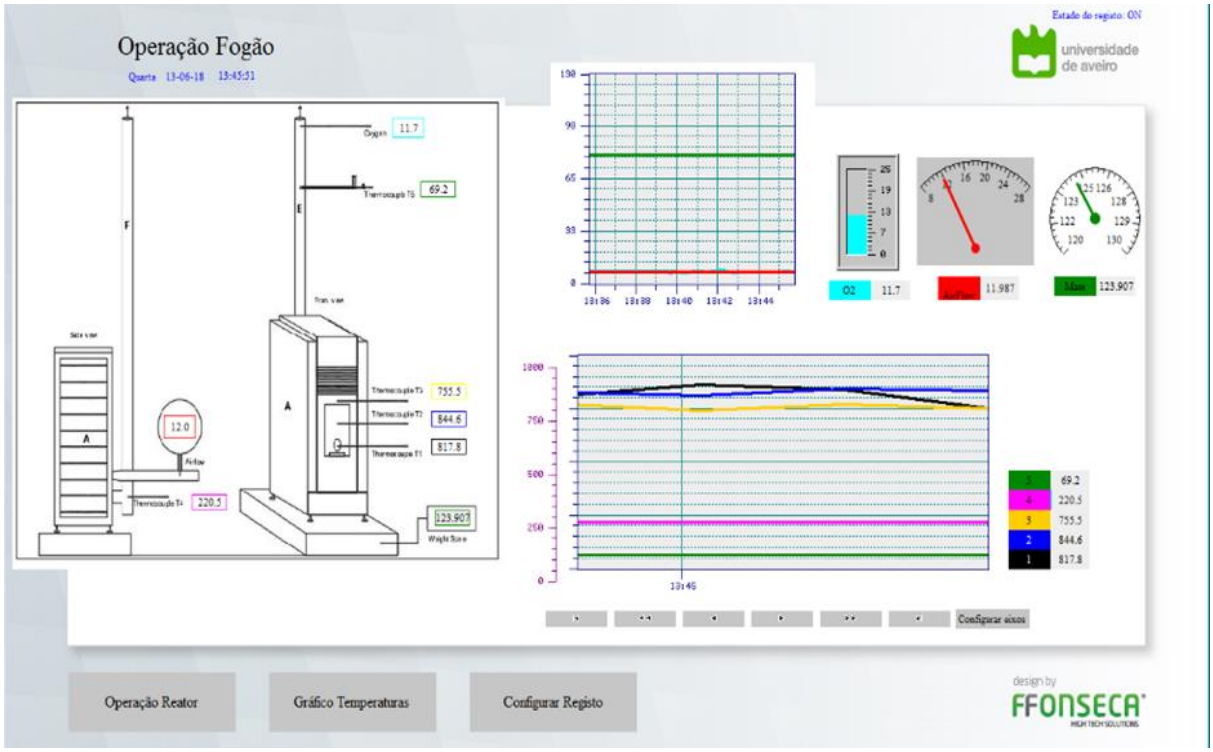

Figure 3.9. Graphic interface for monitoring and control combustion experiments in pellets stove.

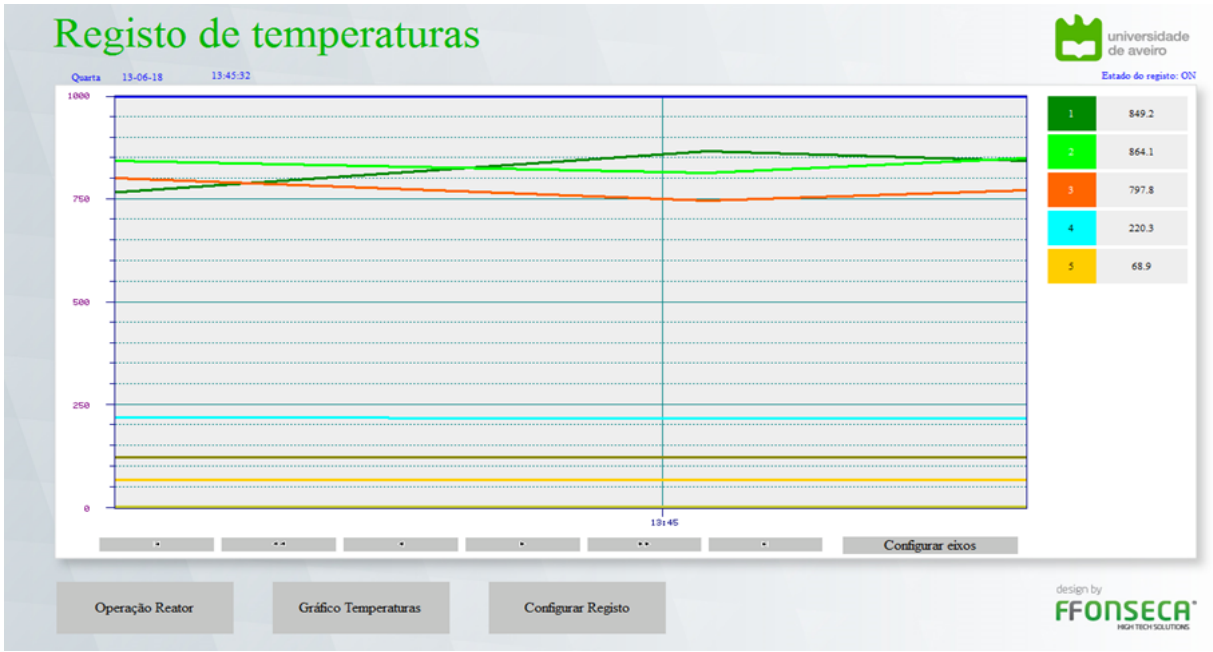

Figure 3.10. Temperature profile real time monitoring. Interface developed in WebAcess software.

3.2 Type biomass pellets

Pelletized woody biomass was used for the combustion tests. Eight types of pellets were used. Pinewells pellets are commercial and certificated being produced by Pinewells S.A. and accomplish the European ENplus-A1 standard. Acacia pellets, and torrified pellets are commercial, non-certified pellets made from different woody biomass. BioSolids are a mixture of biomass from poplar tree fertilized with sewage sludge and cereal straw. Mud pellets are a mixture of poplar tree fertilized with sludge from the wastewater treatment plant of the dairy industry and cereal straw. AF-2 and AF-8 are made of poplar from Italy meanwhile Unal is made of poplar from Spain. It was not possible to get information about torrified pellets at this moment. Biomass pellets elemental analysis, moisture and ash are presented **Table 4**. The elemental analysis was carried out in the laboratory of the Environmental and Planning department at the University of Aveiro **Figure 3.11.** shows the macroscopic aspect of some of the pellets tested.

Figure 3.11. Some samples of pellets tested during combustion experiments. a) BioSolids Af-8 ; b) torrified pellets ; c) Acacia pellets ; d) Pinewells Pellets.

Table 4. Elementary analysis of biomass pellets used in the experiments in dry basis.

Biomass Pellet	Content (%wt, dry basis)					Moisture (%wt wet basis)	LHV kJ/kg
	C	N	H	S	Ash		
Acacia	46.9	0.89	6.1	na	1.62	11.99	17545.52
Pinewells	48.7	0.78	6.1	<0.01	≤ 0.70	9.00	18276.63
BioSolids Unal	48.4	0.6	5.8	0.1	2.5	8.9	17974.2
BioSolids AF-2	48.1	0.7	5.6	0.1	3.1	6.0	17685.14
Mud AF-8	47.9	1.0	5.6	0.1	3.6	8.8	17662.28
Mud Unal	48.2	0.4	5.8	0.1	2.8	8.2	17890.73
BioSolids AF-8	47.9	0.6	5.6	0.1	3.2	6.3	17594.04
Torrified	na	na	na	na	na	3.0	23000.00

na – not available at the moment.

3.3 Pellets stove's set up

A total of eight experiments were conducted to obtain data of the combustion process. The total time per each combustion experiment was 60 minutes. The stove was operated in manual mode. The configuration of manual mode was *Fire 3* and *Fan 3*. The indicator *Fire* sets a rate for feeding the pelletized fuel into the combustion chamber. The stove automatically controls the intake of the primary air inlet. Each of the combustion experiments was performed after a preheating period of the pellet stove. It took about 35 minutes to start the experiments to ensure that the combustion process had already attained a steady operation condition. After every combustion experiment the stove was cleaned. The pellets in the pellet storage tank, the ashes in combustion chamber, and the ashes in the stove grate were removed. The setup of operation parameters of the stove is shown in **Table 5.**

Table 5. Pellets Stove's set up for all the experiments conducted.

Experiment No.	Type of Biomass	Operation Mode		Experiment Reference
		Fire	Fan	
1	Pinewell Pellets	3	3	1-3-3
2	Acacia Pellets	3	3	2-3-3
3	Mud AF-8	3	3	3-3-3
4	BioSolids Unal	3	3	4-3-3
5	BioSolids AF-2	3	3	5-3-3
6	Mud Unal	3	3	6-3-3
7	BioSolids AF-8	3	3	7-3-3
8	Torrified Pellets	3	3	8-3-3

4. Results and discussion

4.1 Stoichiometric oxygen consumption

In or to determine the stoichiometric needs of oxygen (Ws) it assumed that all C oxidized into CO_2; all H oxidized into H_2O; all S oxidized into SO_2. It was considered that the nitrogen in the fuel was not oxidized into NOx; and for mass balance it is considered as released N_2. The complex set of intermediate chemical reactions were not considered. It was considered that the gaseous products do not contain species such as CO, H_2, or hydrocarbons. Stoichiometric needs of oxygen are obtained according to the composition of the fuel, by applying Equation 2.

$$Ws = 32 \times \left[\sum \left(Y_{sj} \frac{W_j}{M_j}\right) - \frac{W_{of}}{32}\right] \quad \text{(Eq.2)}$$

The combustion air needs for is based in the stoichiometric oxygen consumption for each type of pellets, is estimated using Equation 3 [51].

$$w_{sA} = w_s \times \left(1 + 3.76 \times \frac{M_{N_2}}{M_{O_2}} + 7.66 \times W_{V_A} \times \frac{M_{H_2O}}{M_{O_2}}\right) \quad \text{(Eq.3)}$$

4.2 Carbon balance

Carbon balance is a ratio between the amount of carbon leaving the system in flue gases in the form of CO_2, CO, hydrocarbons, TOC, the solid carbon present in the ashes (bottom bed and fly ashes) and the amount of carbon fed in the pellets to the system (the stove). Carbon balance values close to one indicates high combustion efficiency. In this work and considering the goal of estimating the amount of carbon from the fuel converted in gaseous compounds, in the calculation it was not accounted the amount of carbon content remaining in the ash. According to the results for all the types of pellets tested, the overall carbon conversion calculated based on concentrations of CO_2, CO and TOC measured in the combustion flue gas were higher than 50%. Mud-Unal and Pinewells pellets, presented the high conversion of fuel Carbon to gaseous components containing carbon, 78% and 76% respectively. The lowest value of conversion of fuel carbon to gaseous components containing carbon was observed for the BioSolids Unal pellets, that only reached 54% conversion.The results for the experiments conducted in the pellets stove are present in **Table 6.** Carbon balance was calculated using Equations 4, 5 and 6.

$$b = \frac{\dot{m}_{c,out}}{\dot{m}_{c,in}} \quad \text{(Eq.4)}$$

$$\dot{m}_{c,in} = \dot{m}_f \times W_c \times (1 - W_w) \quad \text{(Eq.5)}$$

$$\dot{m}_{c,out} = \dot{m}_{c,CO_2} + \dot{m}_{c,CO} + \dot{m}_{c,TOC} \quad \text{(Eq.6)}$$

Table 6. Carbon balance results for the experiments conducted.

		Biomass pellet type							
Parameter	**Units**	Pinewells	Acacia	Mud Af-8	Mud Unal	BioSolid Unal	BioSolids Af-2	BioSolids AF-8	Torrified
Reference	[-]	1-3-3	2-3-3	3-3-3	6-3-3	4-3-3	5-3-3	7-3-3	8-3-3
Carbon Balance	[-]	0.76	0.64	0.62	0.78	0.54	0.64	0.72	-

4.3 Nitrogen conversion

As mentioned in section 2.6.2, for combustion temperatures lower than 1300 °C the fuel NOx mechanism is the primary response from the formation of NO and NO_2. The temperatures observed in the combustion chamber of the pellet stove during the experiments were much lower than 1000 °C. Therefore, the nitrogen content in the fuel is the main responsible for the formation of NO and NO_2 during the combustion process. The fuel nitrogen conversation to NO is analyzed in section 5.8.3. The biomass conversion to NO and NO_2 was calculated based on the NO and NO_2 concentration measured in the combustion flue gas at the chimney.The Nitrogen conversion results for the experiments conducted are present in **Table 7.** Conversion of biomass Nitrogen to NO showed values between 7% (for Pinewells pellets) to 28%(for Mud Unal pellets). The biomass Nitrogen conversion to NO_2 showed results between 0.01 % from Pinewells Pellets to 0.42% from Mud Af-8.

Table 7. Carbon balance results for the experiments conducted.

		Biomass pellet type							
Parameter	**Units**	Pinewells	Acacia	Mud Af-8	Mud Unal	BioSolid Unal	BioSolids Af-2	BioSolid s AF-8	Torrified
Reference	[-]	1-3-3	2-3-3	3-3-3	6-3-3	4-3-3	5-3-3	7-3-3	8-3-3
Nitrogen Conversion to NO	[-]	0.07	0.11	0.14	0.28	0.14	0.14	0.16	-
Nitrogen Conversion to NO_2	[-]	0.0001	0.0033	0.0042	0.0010	0.0023	0.0003	0.0003	-

4.4 Temperature profiles

The stove was set in for all the experiments in the same operation mode: "Fire 3" and "Fan 3". The characteristics of the biomass pellets were the changing variable. It was observed in the experiments the influence of the pellets characteristics in the combustion conditions such the flue gas temperature at distinct locations along the combustion system. The effect of elemental composition and the physical characteristics (length) of the pellets was also

influencing the steady operation of the stove. Temperatures profiles showed peaks and drops of temperatures, during the combustion experiments.

The highest value of temperature for all the experiments was reached in the thermocouple T1, located in the combustion chamber (see **Figure 3.5**). Commercial torrified pellets reached highest average temperatures in thermocouple T1 during the experiments, 853 ±9.9 °C; also, torrified pellets reached the highest recorded 974 ±11.2 °C (T1 thermocouple). Higher rate of thermal energy production is associated with higher rates of fuel consumption (see section 4.5) as occurred with torrified and pinewells pellets. Acacia pellets showed the lower combustion temperatures and an operation condition with higher excess of air (see **figure 4.1).** In fact, it is recognized that lower temperatures are typically associated with high excess of air, because of higher losses of thermal energy carried out by the combustion flue gases [26].

It was observed that lower temperatures and higher excess of air, were associated with physical characteristics of pellets such; (i) the diameter or the length that not accomplish the stove requirements (see section 3.1), (ii) pellets with high density, or pellets that do not maintain their consistency during the displacement from the pellets store tank, to the combustion chamber and became dust (generate high amount of fine particles such as the non-commercial pellets tested), (iii) chemical composition such high ash and moisture content. The moisture content in the fuel is a detrimental characteristic, because evaporation of water is an endothermic process. Therefore, higher content of moistures in pellets contributed to lower combustion temperatures as was observed with Acacia pellets.

During the combustion experiments, the behavior of temperature in thermocouples T1, T2, T3 located in the combustion chamber (**Figure 3.4**, **3.5**) was characterized by a sudden rise in the first minutes after a load of pellets dropped into the combustion chamber. The rise was followed by a decrease in the temperature. This temperatures fluctuations are visible in the temperatures profiles (Figures **4.2**, **4.9**). The fluctuations were more evident when the pellets had; (i) a smaller length than the required for stove operation (10 to 30 mm), (ii) high content of ash, (iii) high content of moisture, (iv) poor consistency. This four characteristics create some difficulties in supporting the combustion process and consequently the drop in the temperature at the combustion chamber because, the amount of fuel in the combustion chamber is not enough to sustain proper combustion. To compensate for the drop of temperature the stove automatically loaded more fuel into the combustion chamber, achieving a sudden rise of the temperature after a couple of minutes. This period of elevated temperatures is coincident with the existence of a high amount of loaded pellets in the combustion grate and an intense flame in the combustion chamber. The period of temperature drop is coincident with a small flame.

This typical operation pattern of temperature profile fluctuation was observed during the several experiments of combustion of pellets. Even though, this phenomenon was more evident when non-commercial, non-certified pellets were tested **(e.g., Figure 4.3, 4.6)** also commercial pellets also showed this pattern **(Figure 4.2, 4.9)**.

The fluctuation in the temperatures profiles along time, is related to the heterogeneous physical characteristics of the pellets and discontinuous mode of pellet feeding by the screw feeder of the stove. In fact, the feeding system of the stove do not guarantee a constant supply of pellets in the combustion chamber.

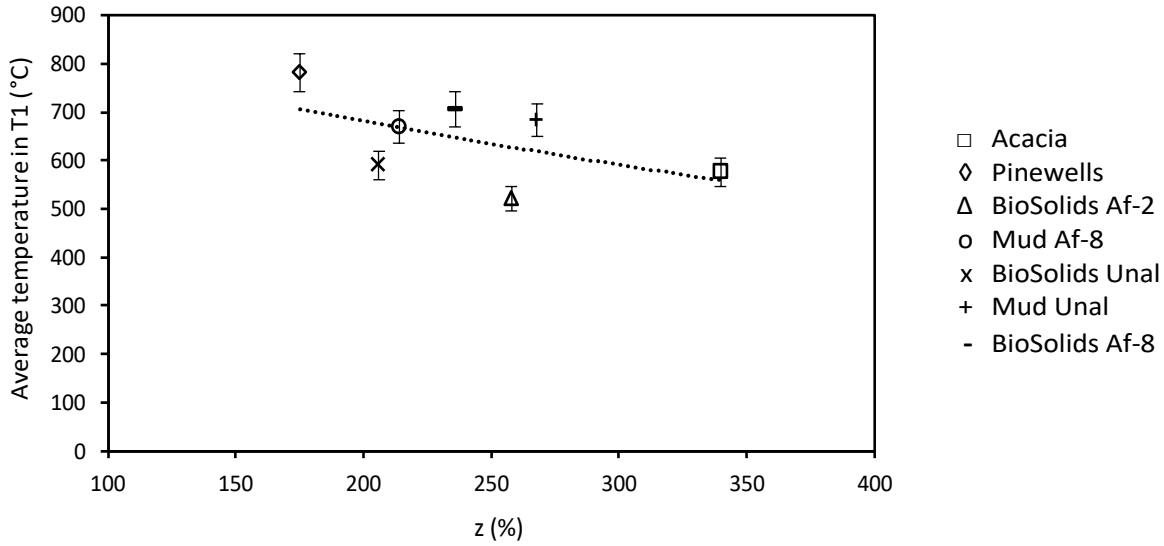

Figure 4.1. Shows the correlation between the primary excess of air and the average temperature reached in thermocouple T1.

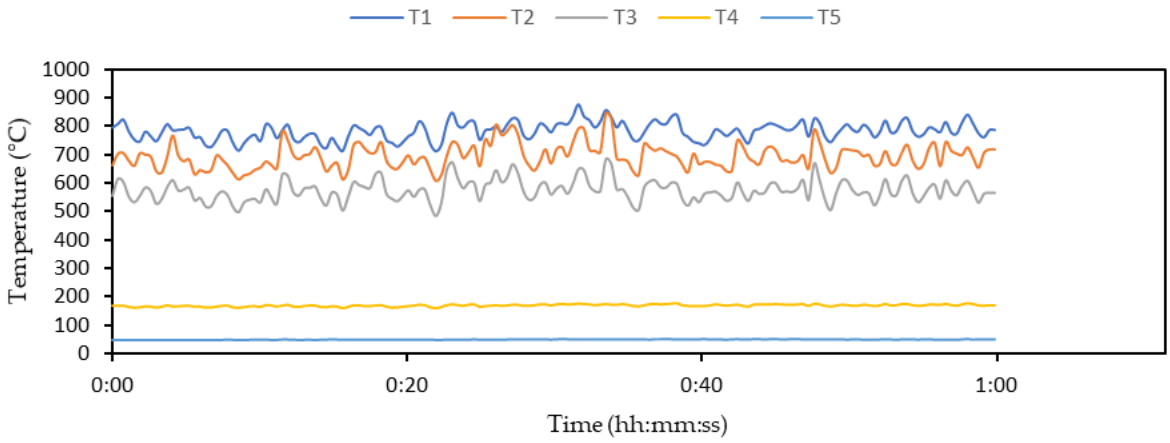

Figure 4.2. Shows the temperature evolution during the combustion of Pinewells pellets. Location of temperature measurement according to Figure 3.4 and 3.5.

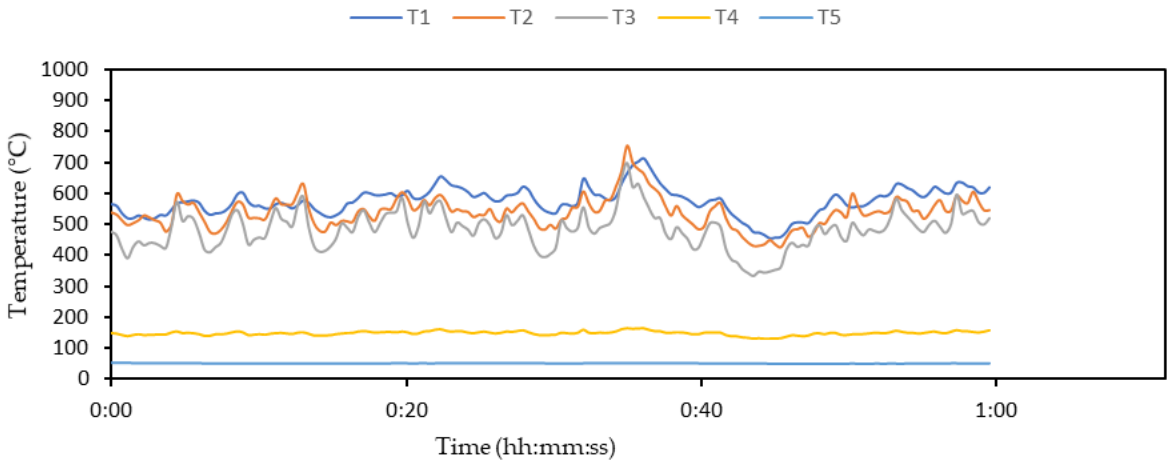

Figure 4.3. Shows the temperature evolution during the combustion of Acacia pellets in the stove. Location of temperature measurement according to Figure 3.4 and 3.5.

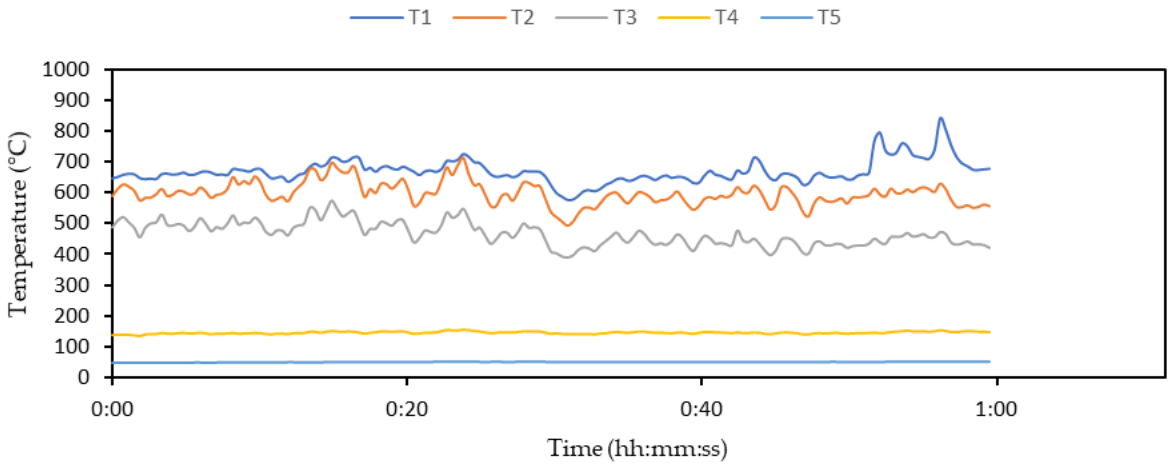

Figure 4.4. Shows the temperature evolution during the combustion of Mud Af-8 pellets in the stove. Location of temperature measurement according to Figure 3.4 and 3.5.

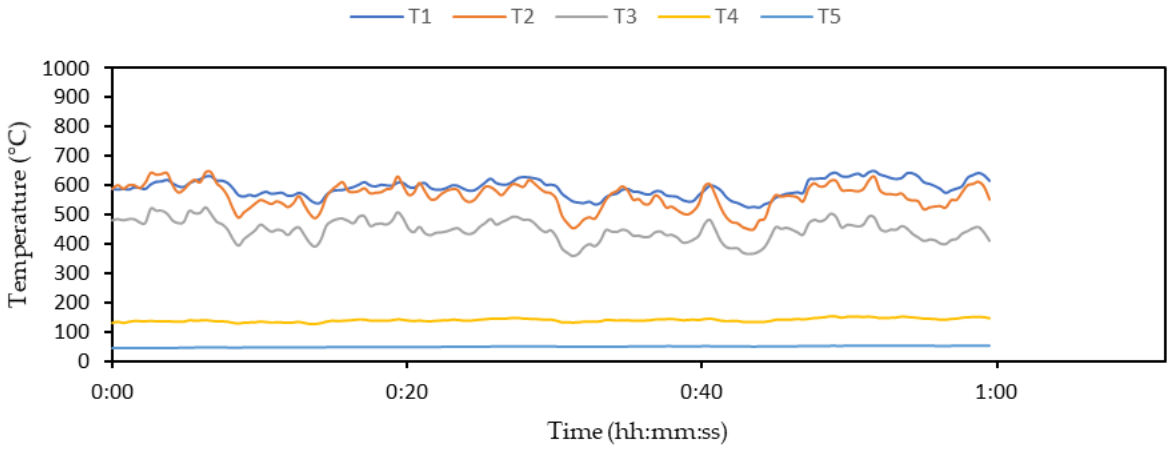

Figure 4.5. Shows the temperature evolution during the combustion of BioSolids Unal pellets in the stove. Location of temperature measurement according to Figure 3.4 and 3.5.

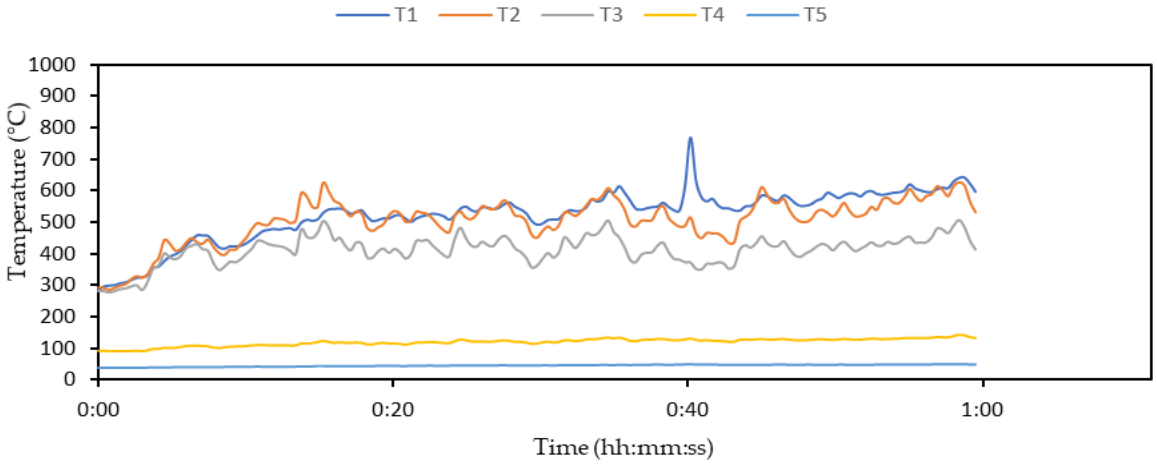

Figure 4.6. Shows the temperature evolution during the combustion of BioSolids AF-2 pellets in the stove. Location of temperature measurement according to Figure 3.4 and 3.5.

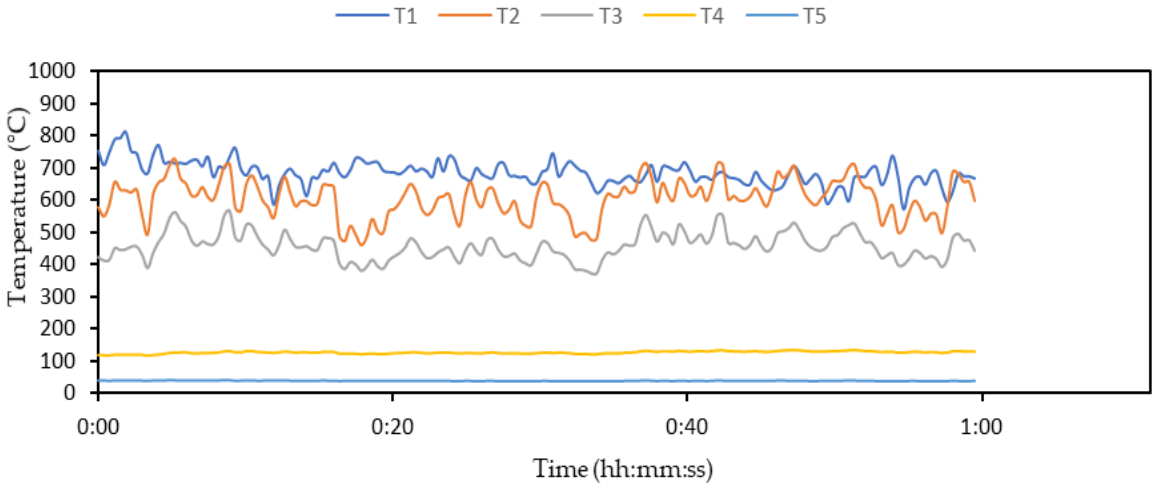

Figure 4.7. Shows the temperature evolution during the combustion of Mud Unal pellets. Location of temperature measurement according to Figure 3.4 and 3.5.

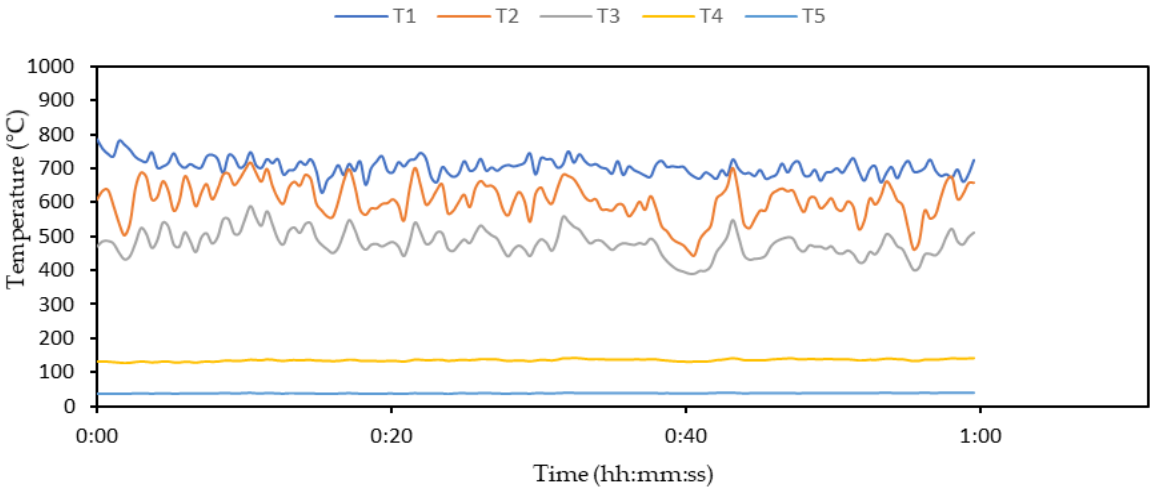

Figure 4.8. Shows the temperature evolution during the combustion of BioSolids AF-8 pellets in the stove. Location of temperature measurement according to Figure 3.4 and 3.5.

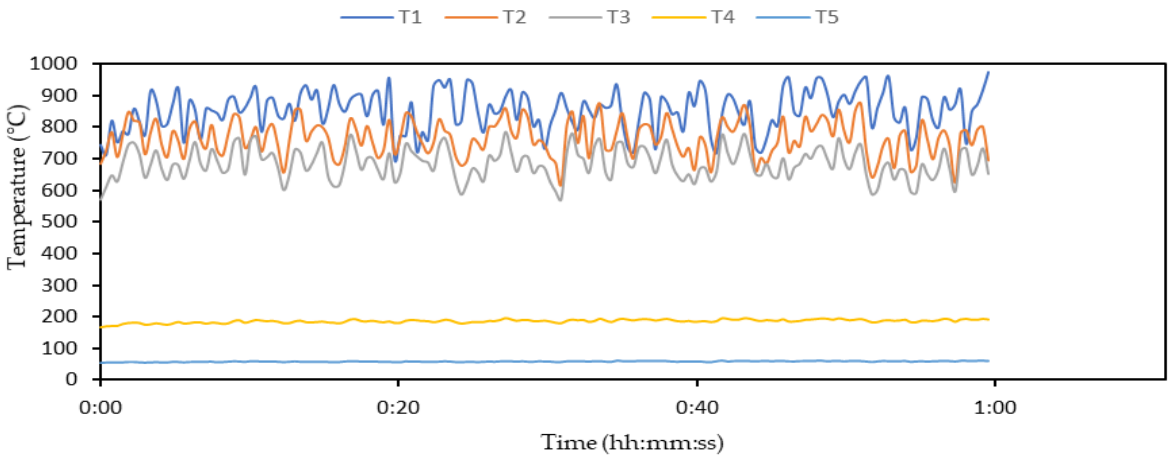

Figure 4.9. Shows the temperature evolution during the combustion of torrified pellets in the stove. Location of temperature measurement according to Figure 3.4 and 3.5.

4.5 Combustion air flow rate and fuel consumption

Figures 4.10 to **4.17** show the evolution of fuel mass in the fuel reservoir of pellets stove the primary air flow rate entering the combustion chamber during the combustion experiments, and the fuel consumption along the time. The evolution of the fuel mass in the fuel reservoir was used to calculate the average fuel consumption rate (**Figure 4.18)**.

The temperature at the combustion chamber reflects the fuel consumption rate. Torrified pellets reached the maximum fuel consumption 1.40 $kg \cdot h^{-1}$, (see **Figures 4.17** and **4.18),** followed by Pinewells 1.10 $kg \cdot h^{-1}$, this promoted the higher temperatures observed in the combustion chamber. The average primary air intake flow was 12.78 $Nm^3 \cdot h^{-1}$ for all experiments conducted.

In general, the behavior of the mass of the fuel in the stove showed a decreasing linear tendency. The variations observed in the decreasing linear tendency and in the fuel consumption along the time, can be associated to some fluctuations in the pellets feeding due to the screw type feeding of the stove and because to the heterogeneity of the physical properties of the pellets. The evolution of the mass of the fuel on the stove depends on the fuel's physical and chemical characteristics. Torrified pellets showed the steadiest mass's evolution of the stove, and perhaps that can be related to better physical properties of the pellets in result of the torrification process.

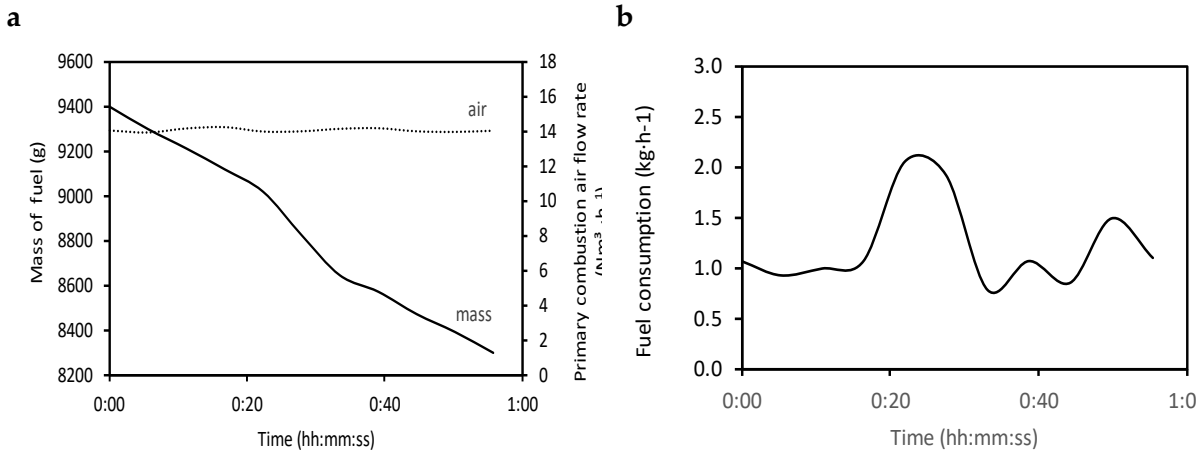

Figure 4.10. During the combustion of Pinewells pellets. a) Combustion air flow rate and mass of biomass; b) Fuel consumption along the time.

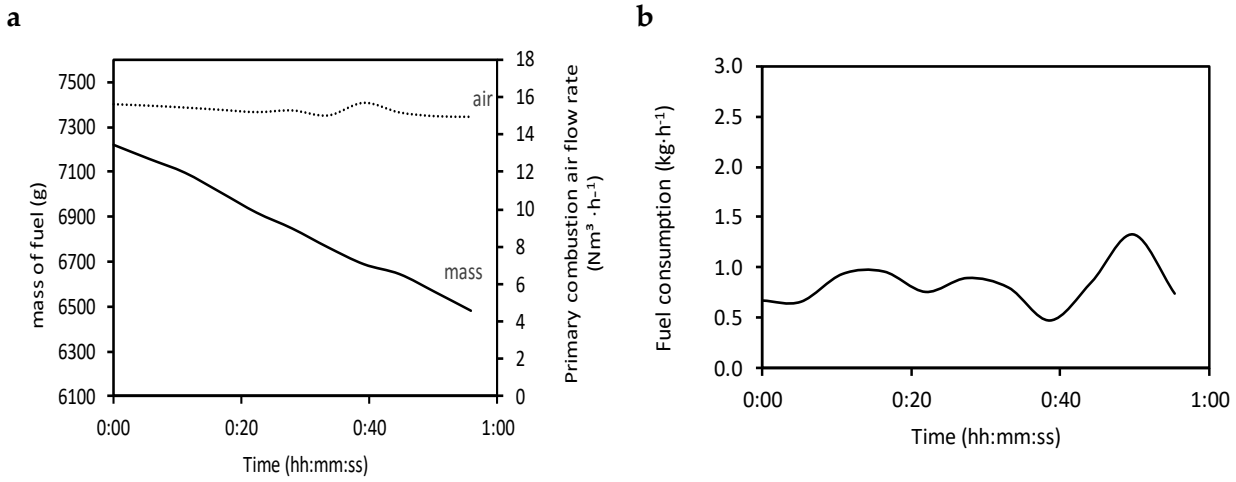

Figure 4.11. During the combustion of Acacia pellets. a) Combustion air flow rate and mass of biomass fuel; b) Fuel consumption along the time.

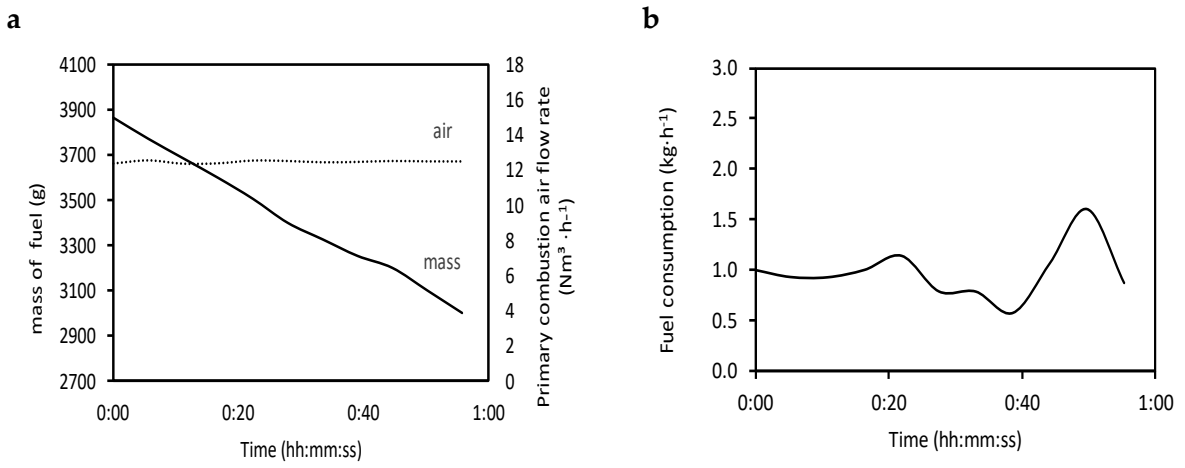

Figure 4.12. During the combustion of Mud Af-8 pellets. a) Combustion air flow rate and mass of biomass fuel; b) Fuel consumption along the time.

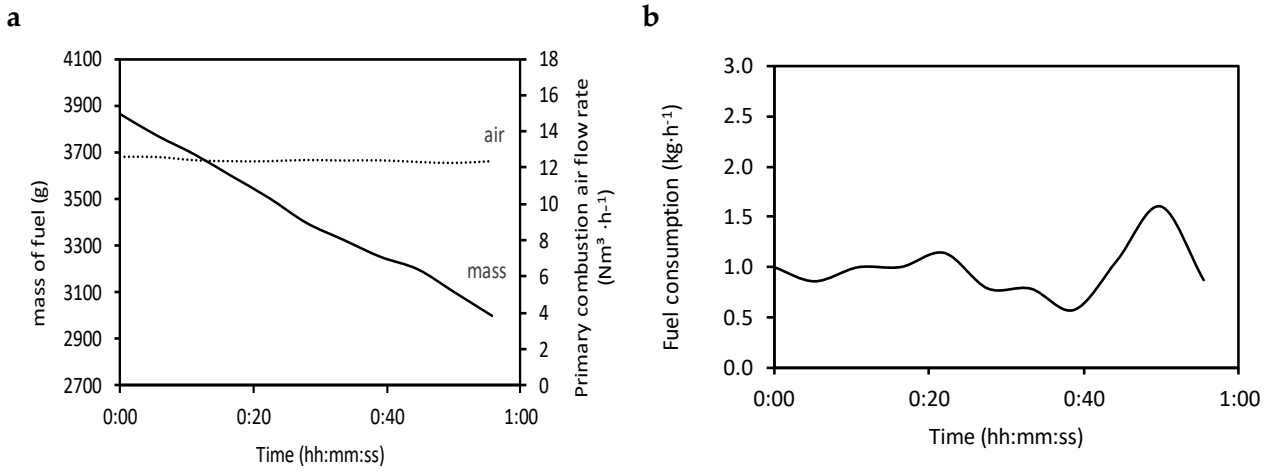

Figure 4.13. During the combustion of BioSolids Unal pellets. a) Combustion air flow rate and mass of biomass fuel; b) Fuel consumption along the time.

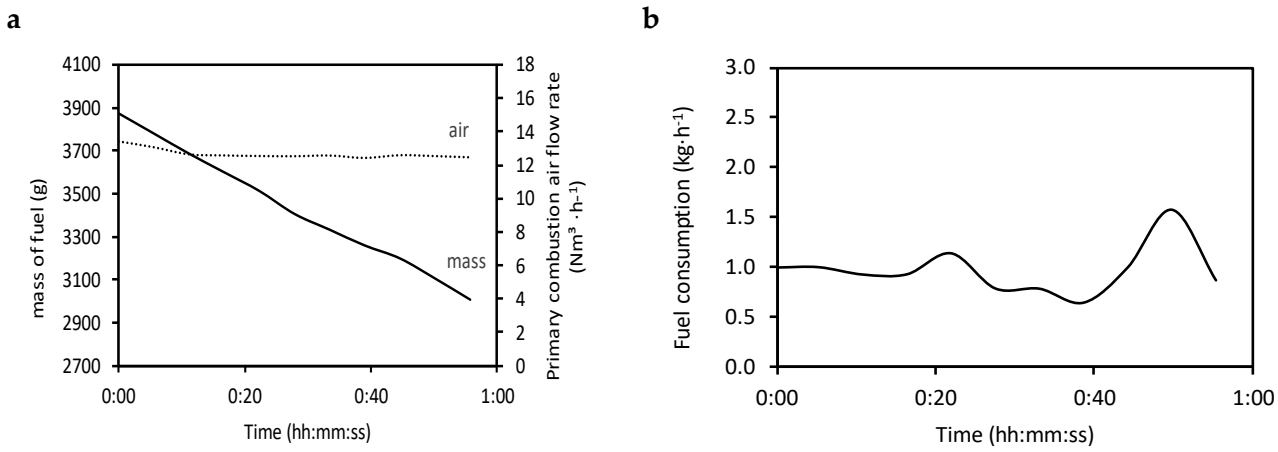

Figure 4.14. During the combustion of BioSolids AF-2 pellets. a) Combustion air flow rate and mass of biomass fuel; b) Fuel consumption along the time.

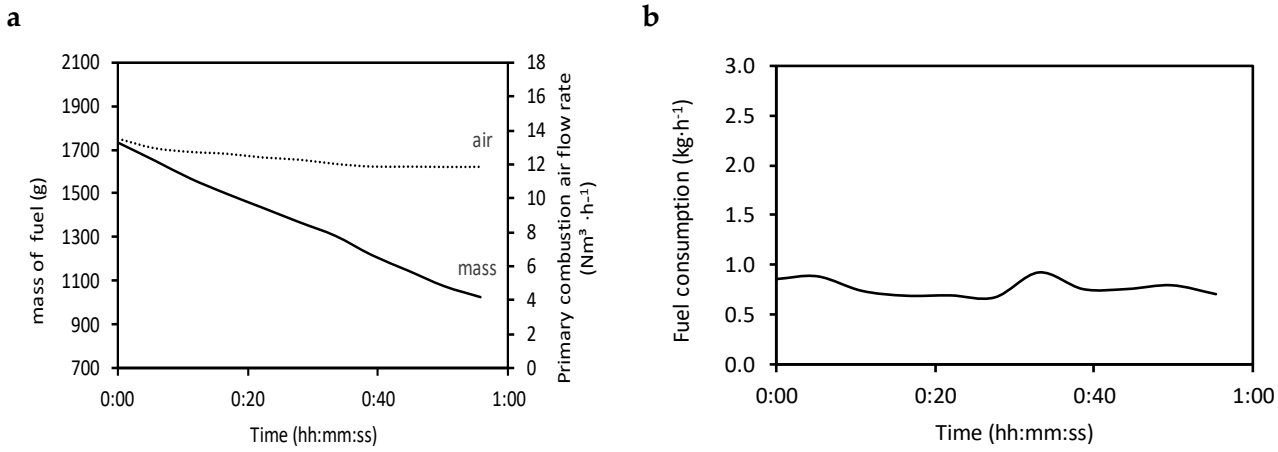

Figure 4.15. During the combustion of Mud Unal pellets. a) Combustion air flow rate and mass of biomass fuel; b) Fuel consumption along the time.

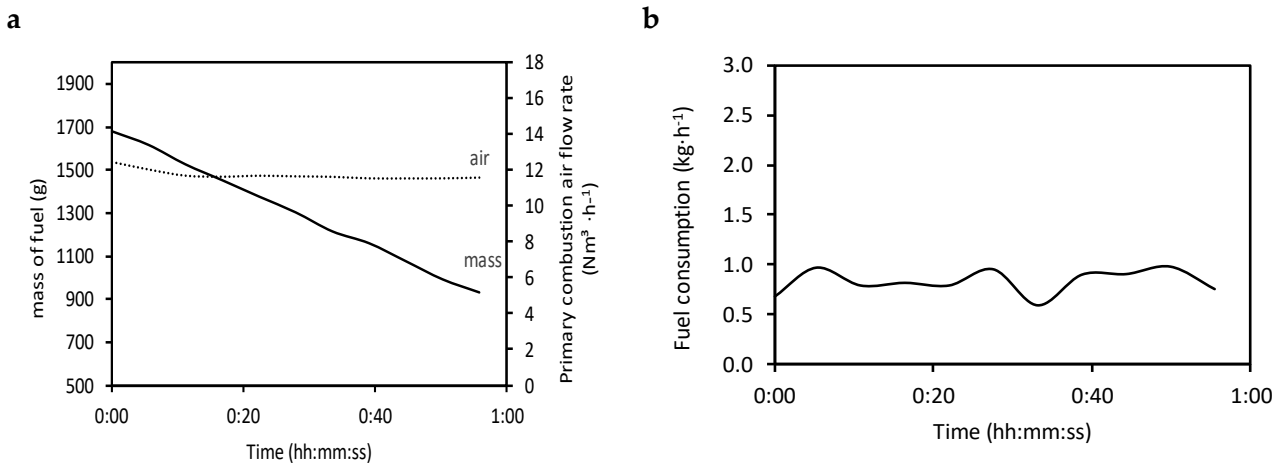

Figure 4.16. During the combustion of BioSolids AF-8 pellets. a) Combustion air flow rate and mass of biomass fuel; b) Fuel consumption along the time.

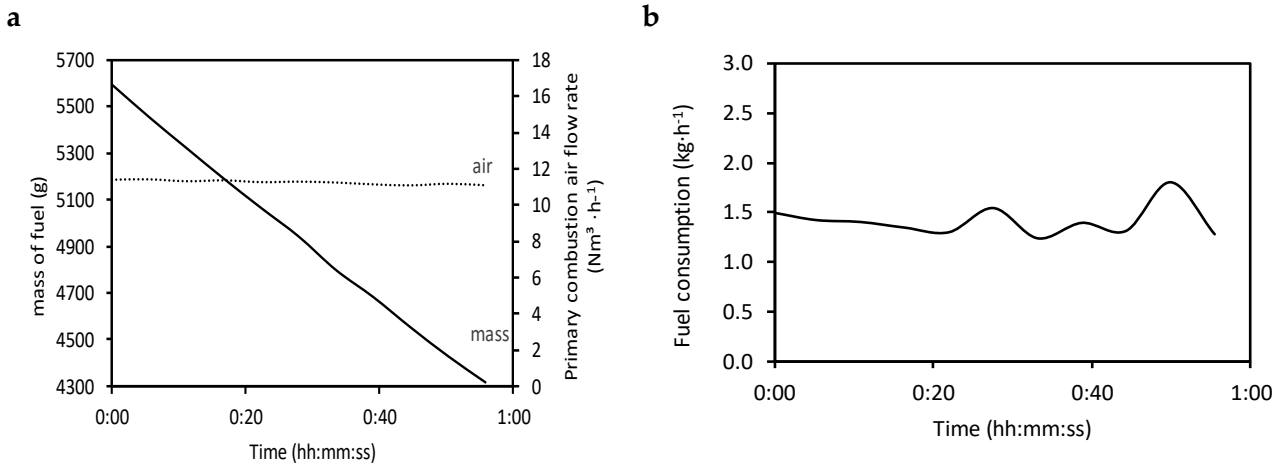

Figure 4.17. During the combustion of Torrified pellets.) Combustion air flow rate and mass of biomass fuel; b) Fuel consumption along the time.

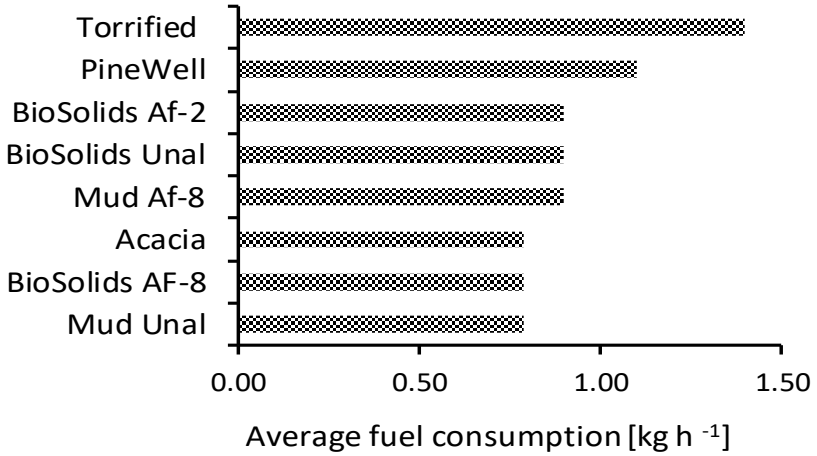

Figure 4.18. Average fuel consumption of the different biomass pellets during the combustion experiments in the stove.

4.6 Excess of air

As it was mentioned before the characteristics of biomass pellets influenced the operating conditions. Operating variables as the stoichiometry, excess of air, fuel consumption, temperature in the combustion chamber, temperature in the exit of the chimney, carbon balance, nitrogen conversion was influenced by the characteristics of the biomass pellets. The excess of air is an important operating characteristic of domestic combustion technology. Domestic systems usually operate with high values of excess of air (e.g., $\lambda > 2$ or z >200%) to reduce CO emission. In order to determinate the excess of air, the stoichiometric of oxygen consumption according the composition of the fuel was calculated using Equation 2 and 3. The excess of air was calculated using Equation 7 and Equation 8 for the distinct biomass pellets. For torrified pellets, it was no possible to obtain information about elemental composition, and therefore, the stoichiometric air needs, primary air excess, primary excess air ratio, carbon balance and nitrogen conversion were not calculated.

$$z = \frac{\dot{m}_{air}}{W_{SA} \times \dot{m}_f} \times 100 \qquad \text{(Eq.7)}$$

$$\lambda = \frac{z}{100} + 1 \qquad \text{(Eq.8)}$$

Stoichiometric air needs for the several types of pellets was is a range between 4.4 to 4.6 $Nm^3 \cdot kg^{-1}$, due to the similar elemental composition of the distinct pellets types. The higher primary excess air value used during the combustion was z =340%, during the combustion of Acacia pellets, and the lower primary excess air was z = 175%, during the combustion of Pinewells pellets. The primary air flow was automatically vented by the stove through an electric fan located downstream the combustion chamber. As a user of this technology, we have no control over this variable. **Table 8** resume all the parameters related to operation conditions.

Table 8. Resume all the parameters related to operation conditions.

Parameter	Units	Biomass pellet type							
		Pinewells	Acacia	Mud Af-8	Mud Unal	BioSolid Unal	BioSolids Af-2	BioSolids AF-8	Torrified
Reference	[-]	1-3-3	2-3-3	3-3-3	6-3-3	4-3-3	5-3-3	7-3-3	8-3-3
Fuel Consumption	[$kg_{fuel\ as}$· h^{-1}]	1.10	0.79	0.90	0.75	0.90	0.90	0.79	1.40
Primary Air mass flow rate	[$Nm^3 \cdot h^{-1}$]	14.0 ±0.05	15.3 ±0.07	12.5 ±0.04	12.4 ±0.08	12.4 ±0.03	12.7 ±0.06	11.7 ±0.05	11.2 ±0.04
Stoichiometric combustion air	[$Nm^3_{air} \cdot kg^{-1}$ $_{fuel\ db}$]	4.6	4.4	4.4	4.4	4.5	4.4	4.4	-
Z (primary excess air)	[%]	175	340	214	268	206	258	236	-
λ (primary excess air ratio)	[-]	2.75	4.40	3.14	3.68	3.06	3.58	3.36	-
Combustion Chamber Average Temperature (T1)	[°C]	782 ± 4.7	576 ±7.0	669 ±5.9	684 ±5.9	590 ±4.6	521 ±12.5	706 ±3.8	853 ±9.9
Carbon Balance	[-]	0.76	0.64	0.62	0.78	0.54	0.64	0.72	-
Nitrogen Conversion to NO	[-]	0.07	0.11	0.14	0.28	0.14	0.14	0.16	-
Nitrogen Conversion to NO_2	[-]	0.0001	0.0033	0.0042	0.0010	0.0023	0.0003	0.0003	-

4.7 Influence of fuel ash content in the stove operation

The ash content in the biomass pellets caused operating problems into the stove. Pellets with a content of more than 2.5 %wt ash dry basis, as Mud Af-8, BioSolids Unal, Biosolids Af-2 showed operating problems. During the combustion process, the ash released remains accumulated in the grate of the stove, causing also some locking of the primary air injection holes. For some pellets, the ash particles did not fall to the ash pan because its size was bigger than the holes in the grate. After 60 minutes of continuous operation, the ash blocked the exit of the pellets feeding port. The ash accumulation in the grate of the stove disturbed the proper circulation of the primary air flow through the fuel bed, which has an adverse effect in oxidation of the gaseous compounds. **Figure 4.19** shows an example of ash accumulation in the grate.

Figure 4.19. MUD Af-8 pellets with an ash content of 3.6%wt (dry basis) remains in the combustion chamber blocking the pellets feeding tunnel, after 60 min of operation.

4.8 Combustion flue gas composition

4.8.1 CO, CO_2, O_2

The concentration of CO_2 and CO in the combustion flue gas was directly related to pellets characteristics, temperature and excess of air. Even using high-quality, and certified pellets as Pinewells pellets, wich accomplish the European standard ENplus-A1, it was observed a behavior characterized by fluctuations in the concentration of CO, CO_2, O_2. This behavior is shown in **Figures 4.20** to **4.27** for CO, CO_2, and O_2 concentrations in the exit flue gases along the time, during the combustion experiments conducted.

Mud Af-8 pellets showed fewer variations between lowest and highest concentration level of CO and CO_2 reached (**Figure 4.22**). The CO concentration from BioSolids AF-8 (**Figure 4.26)** shows the highest values 2271 ppm. All the pellets overpassed in some point the 1000 ppm of CO concentration in flue gases. The pattern showed that CO_2 concentration increased with the decrease of O_2 concentration in the flue gases, thus reflecting the lower stoichiometric air ratio operation with the decreasing the O_2 concentration. The behavior of O_2 and CO_2 were complementary during the combustion process. The O_2 concentration in flue gases during the combustion of torrified pellets was the lowest, thus it was expected to be found in torrified pellets the higher values of CO_2 concentration. Torrified pellets reached the highest value of CO_2 concentration in flue gases, around 7.60 vol-% $_{db}$ (**Figure 4.27**).

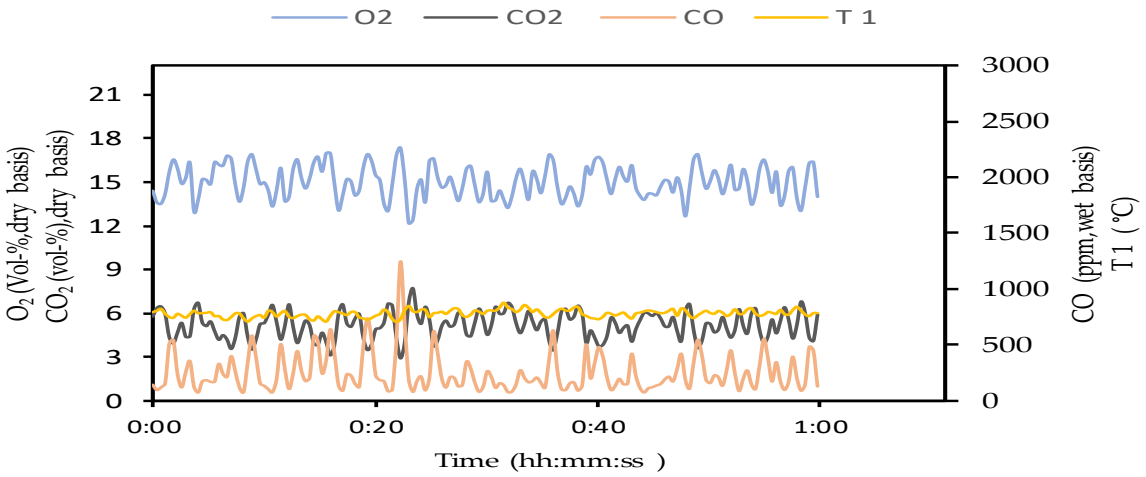

Figure 4.20. CO, CO_2 and O_2 concentrations in the exit flue gases along time during Pinewells pellets combustion.

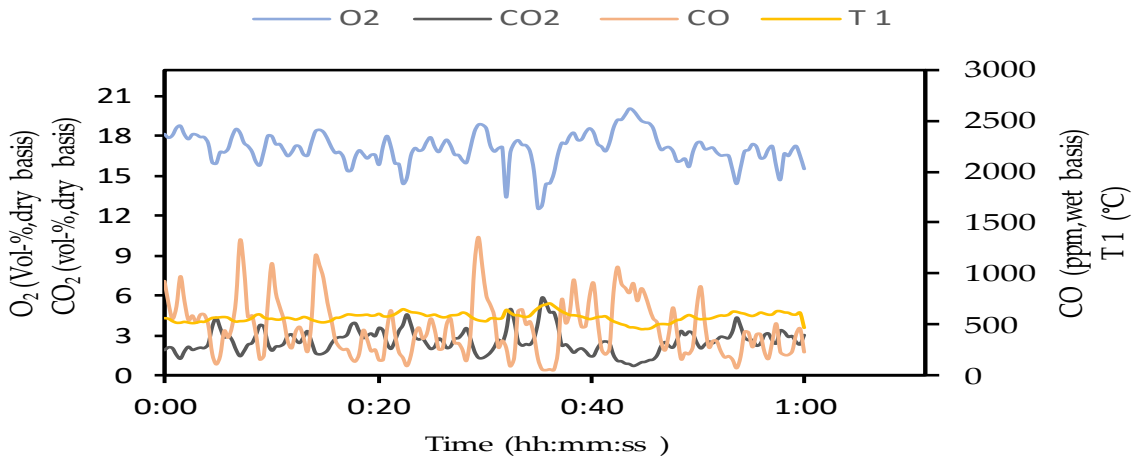

Figure 4.21. CO, CO_2 and O_2 concentrations in the exit flue gases along time during Acacia pellets combustion.

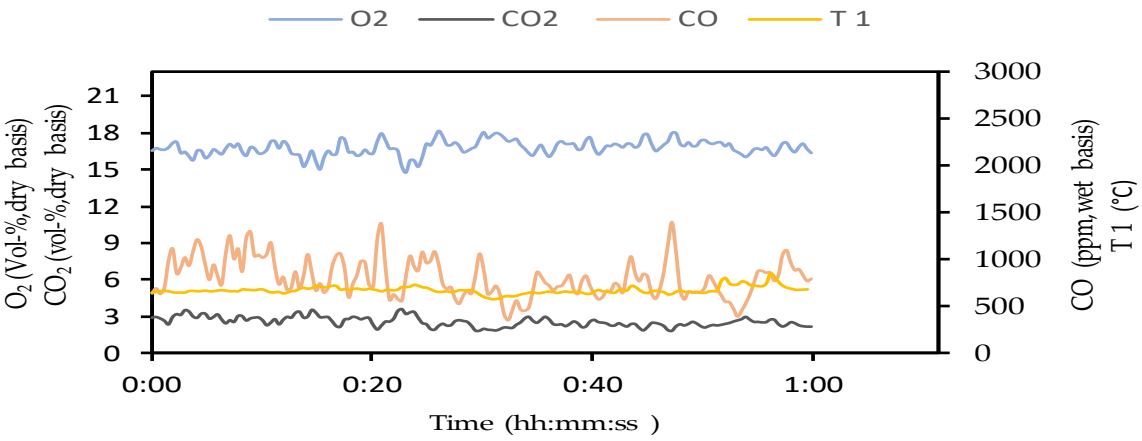

Figure 4.22. CO, CO_2 and O_2 concentrations in the exit flue gases along time during Mud Af-8 pellets combustion.

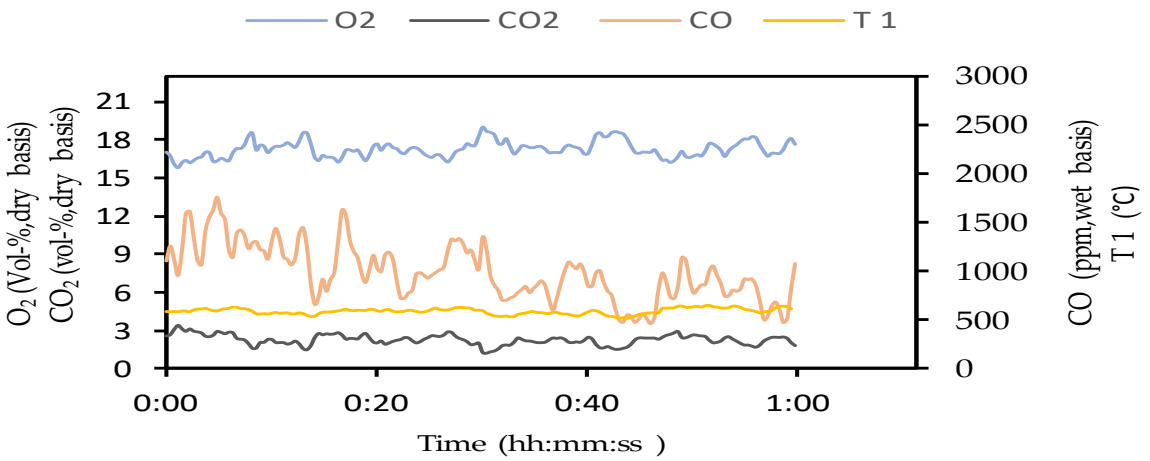

Figure 4.23. CO, CO_2 and O_2 concentrations in the exit flue gases along time during BioSolids Unal pellets combustion.

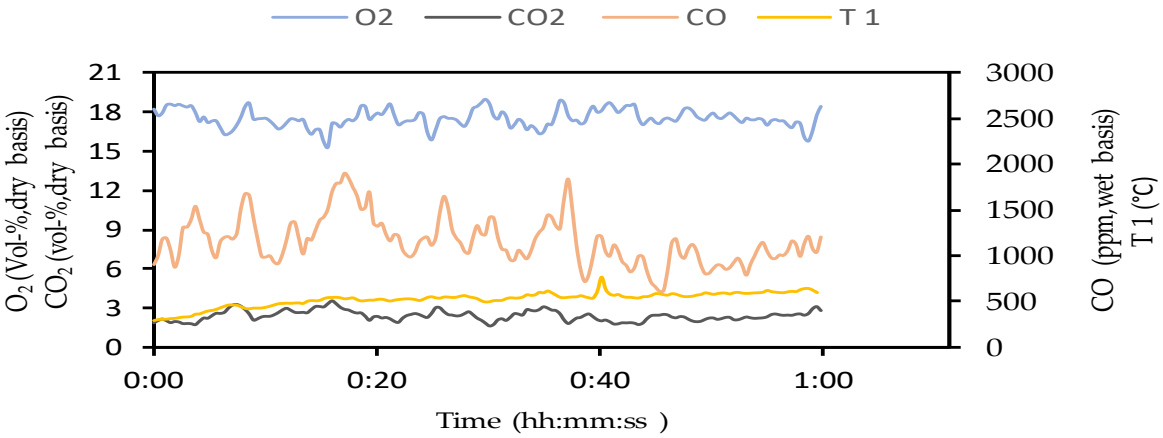

Figure 4.24. CO, CO_2 and O_2 concentrations in the exit flue gases along time during BioSolids AF-2 pellets combustion.

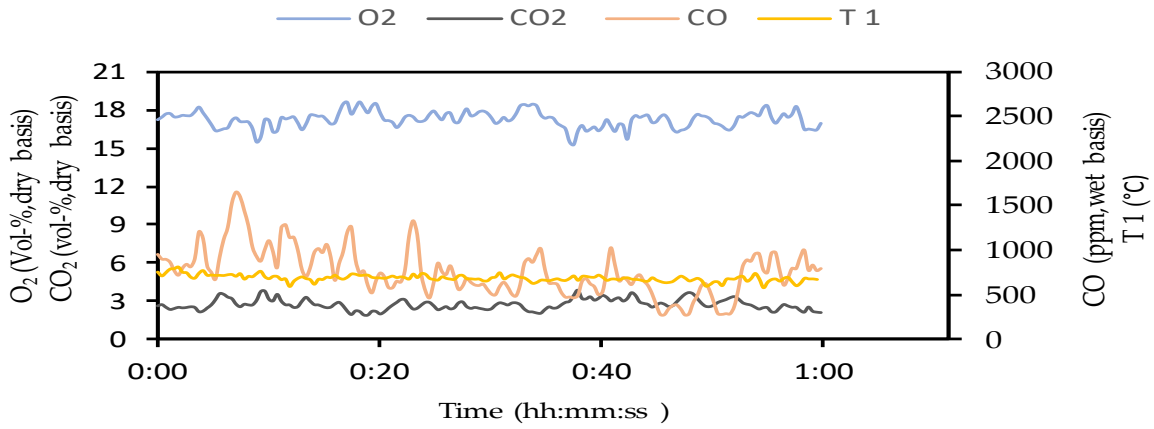

Figure 4.25. CO, CO_2 and O_2 concentrations in the exit flue gases along time during Mud Unal pellets combustion.

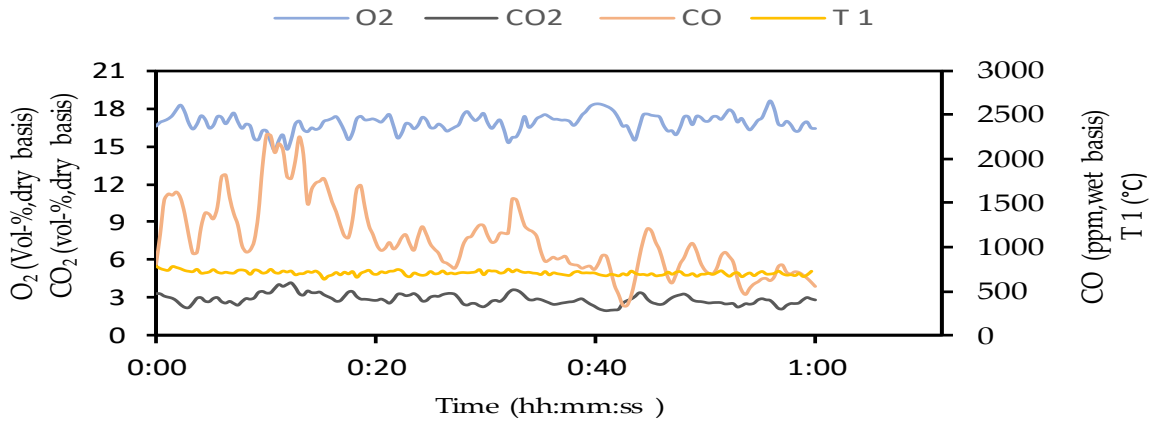

Figure 4.26. CO, CO_2 and O_2 concentrations in the exit flue gases along time during BioSolids AF-8 pellets combustion.

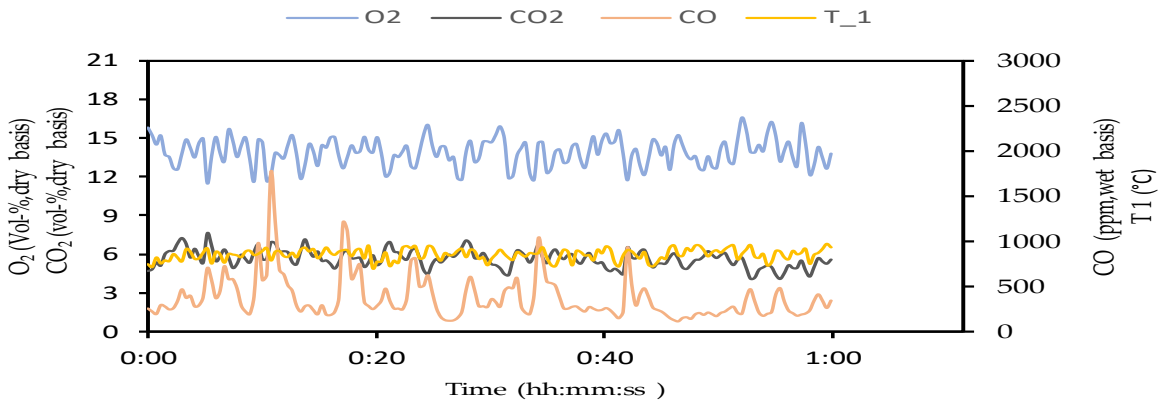

Figure 4.27. CO, CO_2 and O_2 concentrations in the exit flue gases along time during torrified pellets combustion.

4.8.1.1 CO emissions as function of temperature

The concentration CO_2 in flues gases is an indicator of complete combustion, meanwhile the presence of CO, and HC is an indicator of incomplete combustion. A direct correlation exists between incomplete combustion and too low combustion temperatures. Higher temperatures lead to high conversion of fuel Carbon to CO_2, therefore, a low CO emission. The experimental result show that higher temperatures and lower emission of CO were obtained during pinewells and torrified pellets combustion (**Figure 4.28**). In cases in wich sufficient oxygen is available, the lower temperature, is the most critical variable wich leads to an increase in CO emission.

A factor that has an incidence in the reducing of the possible maximum combustion temperature is the content of moisture of the fuel. Acacia pellets had highest content of moisture 11%wt (wet basis), consequently reached the lowest temperatures. However, it did not show the worse CO concentration (**Figure 4.28**). BioSolids Af-2 pellets with a moisture content of 6%wt (wet basis), or BioSolids Unal, or BioSolids Af-8 with moisture content (8.9%wt and 6.3%wt wet basis, respectively) present higher concentration of CO in the combustion flue gases. Other factors such as the ash content should be considered as indirect variable, influencing the temperature in the combustion chamber and related emission of unburned gaseous compounds.

Portuguese regulation sets the emission at a maximum limit of 500 $mg \cdot Nm^{-3}$ corrected to reference value of 11%-vol O_2 and dry gas for biomass combustion [86]. The calculation of the limit emission was based in the average concentration of CO in the combustion flue gases, during the combustion period.

None of the pellets studied here complied the regulation for CO emission (**Figure 4.28**). Emission of CO from BioSolids Af-2 pellets was 7.5 times higher than the limit, meanwhile BioSolids Unal and BioSolids Af-8, reached values of six times higher. Better results of CO emission were obtained with pinewells pellets and torrified, 547 and 615 $mgCO \cdot Nm^{-3}$ (dry gases,11%vO_2) respectively.

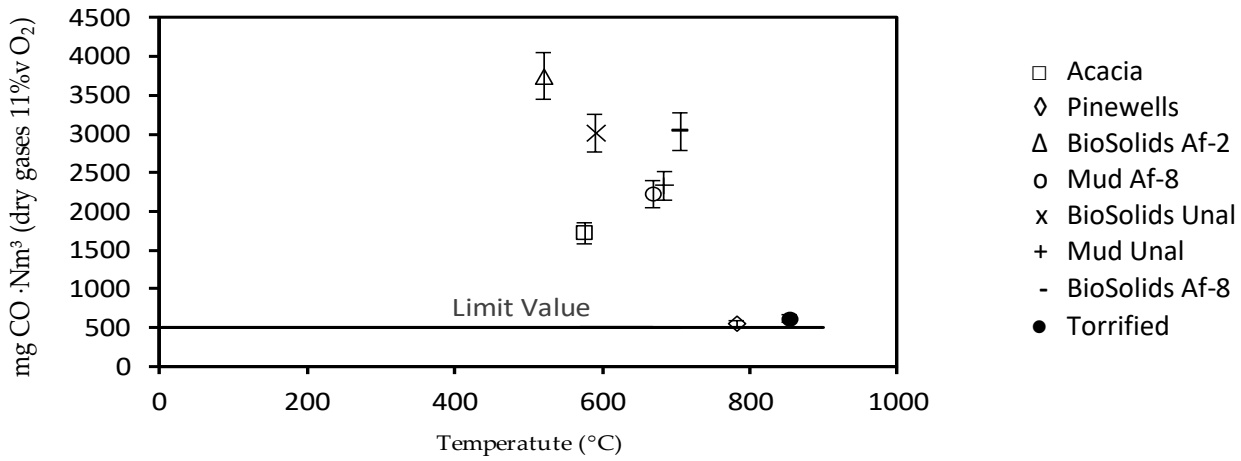

Figure 4.28. CO concentration in combustion flue gas in (mg·Nm^{-3}) as a function of average combustion temperature in thermocouple T1 **(Figure 3.4, 3.5**). CO emission was corrected to reference value of 11%-vol O_2 and dry gas, follow Portuguese regulation [86].

4.8.1.2 CO emission as function of primary excess air ratio

Inadequate mixing of combustion air and fuel in the combustion chamber, or a lack of available oxygen, are factors that favor incomplete combustion conditions. Every biomass pellets have a specific primary air excess ratio (λ), where the CO emissions are minimal; below and above this specific excess air ratio, the CO emissions increase [22]. Also, a higher content of moisture in the pellets and a low fuel consumption usually increase the optimum excess air ratio, and vice versa, promoting incomplete combustion conditions.

This operating characteristics was observed during the combustion of Acacia pellets (11 %wt moisture), and BioSolids Unal (8.9 %wt moisture) (see **Figure 4.29**). These pellets presented an operation condition with high primary air excess ratio (λ), compared with other pellets (see **Table 4** and **Table 8**). For all the experiments it was observed that the primary air excess ratio was between λ=2.75 and λ=4.40 (**Figure 4.29**), that means an excess of primary air between 175 % to 340%. **Figure 4.29** show the correlation between CO emission and primary excess air ratio, showing a tendency for an increase in CO concentration in combustion flue gases with increasing primary air excess ratio (λ).

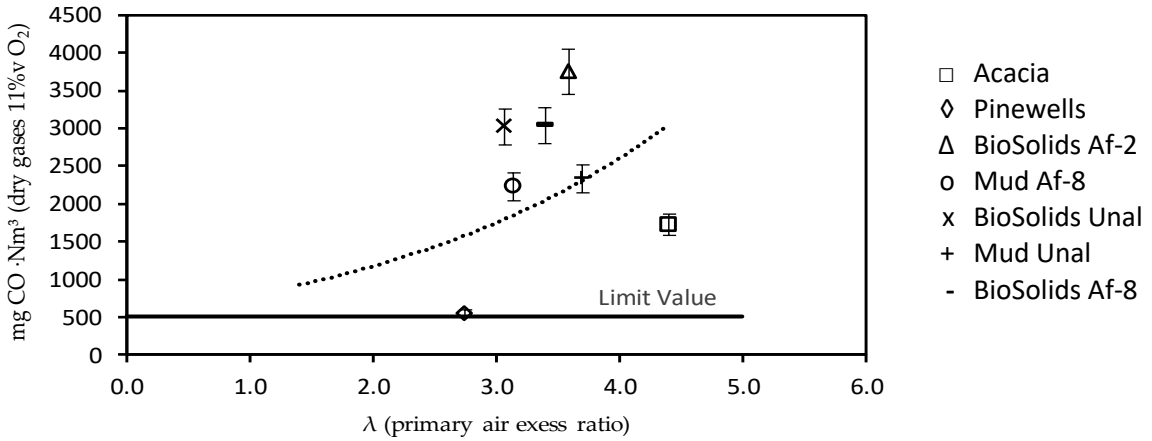

Figure 4.29. CO concentrations in combustion flue gases (in mg·Nm^{-3}) as a function of primary excess air ratio (λ). CO emission are corrected to reference value of 11%-vol O_2 and dry gas, follow Portuguese regulation[86].

4.8.2 CH_4, C_2H_6 and HCl concentration in combustion flue gases

The presence of light hydrocarbons (e.g., methane and ethane) in the combustion flue gases, have a direct relation with incomplete combustion. Ethane and methane are intermediate compounds in the conversion of the Carbon content in the fuel into CO_2. Factors as low combustion temperatures, too short residence times, or lack of available oxygen promote the presence of CH_4 and C_2H_6 in the combustion flue gases. Higher concentration of ethane and methane in flue gases are indicative of lower efficiency in fuel Carbon conversion to CO_2 as occurred during the combustion of Acacia, or BioSolids AF-2 and AF-8 pellets (see **Figures 4.31, 4.34 and 4.36**). It was observed that the CH_4 concentration profile along the time followed a similar pattern as the CO concentration for all the experiments conducted (see **Figures 4.20 to 4.27** and **Figures 4.30 to 4.37**). This can be explained because both of this species are released during the devolatilization step of the biomass combustion. The factors such as for example the low temperature and high primary excess ratio, which promoted the higher concentrations CO, also influenced at the higher concentrations of CH_4 in combustion flue gases at it was observed.

The HCl concentration in combustion flue gases is related with the content of chlorine in the fuel. The HCl concentration in flue gases during combustion of pellets made from wood is expected to be relatively low because of the typical low chorine content in the wood [22]. Pellets made from wood species as Pinewells or Acacia followed this statement and showed a very low concentration of HCl (see **figures 4.30** and **4.31**). However higher emission was observed during BioSolids Af-2, BioSolids Af-8 pellets combustion (see **Figures 4.34** and **4.37**) These pellets were made from a mixture of woody biomass from poplar tree fertilized with sewage sludge and cereal straw, so it can be possible that the straw and the sewage were the contributors for the chlorine and promote a higher HCl concentration in flue gases. Nevertheless, a deeper chemical analysis of these pellets must be made in order to confirm the higher concentration of chorine. Torrified pellets also presented a higher concentration value of HCl (**Figure 4.37**) relative to the others pellet tested. The HCl concentration during the combustion of torrified pellets was 0.33 ppm , which represents a concentration 2.2 times higher than the one obtained with commercial pinewells pellets. Unfortunate it was not

possible to obtain information as elemental composition of torrified pellets in order to know the content of chlorine. **Figures 4-30 to 4-37** show CH_4, C_2H_6, HCl concentration in the combustion flue gases along time.

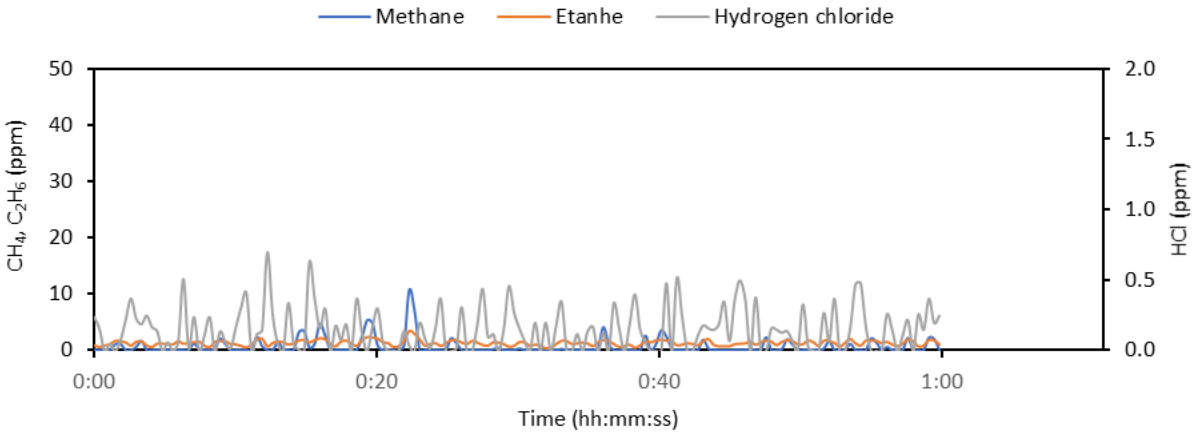

Figure 4.30. CH_4, C_2H_6, HCl concentrations in the exit flue gases along time, during combustion of Pinewells pellets.

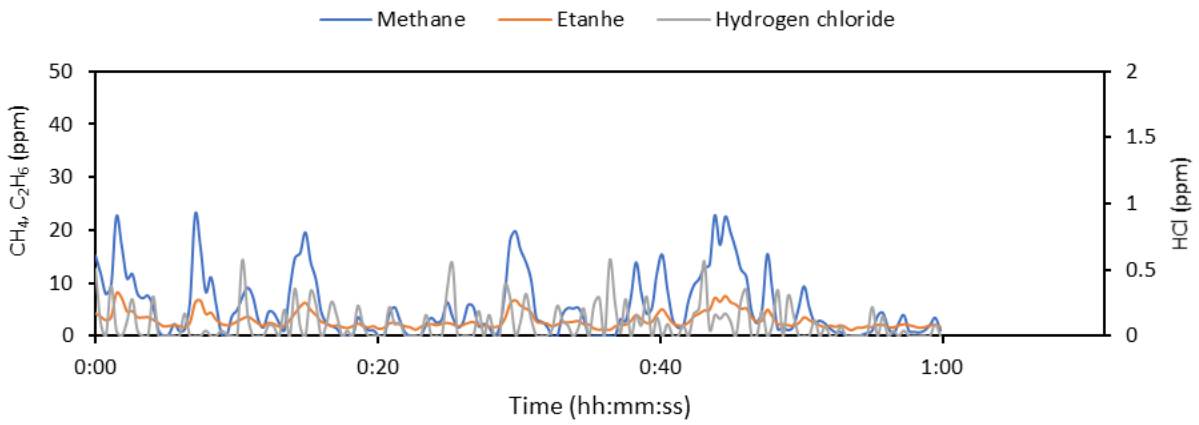

Figure 4.31. CH_4, C_2H_6, HCl concentrations in the exit flue gases along time, during combustion of Acacia pellets.

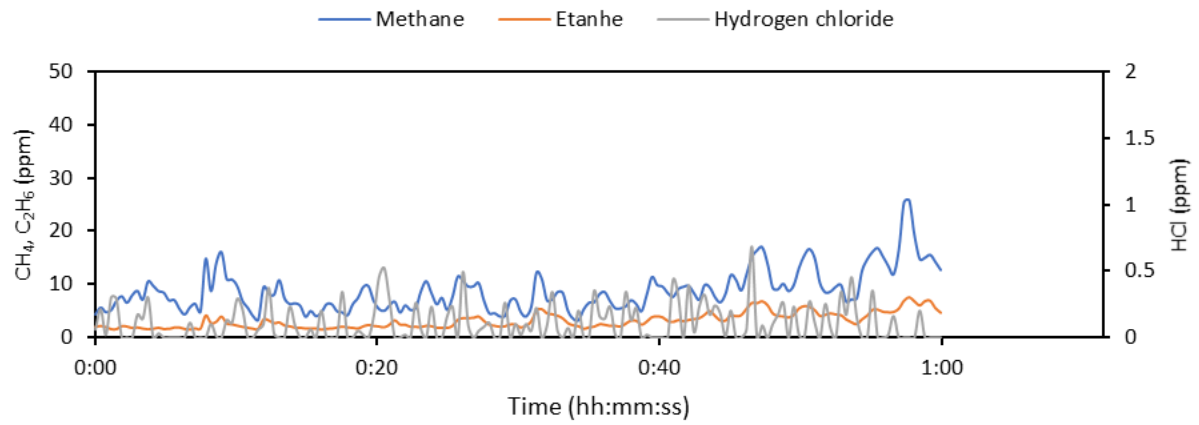

Figure 4.32. CH_4, C_2H_6, HCl concentrations in the exit flue gases along time, during combustion of Mud Af-8.

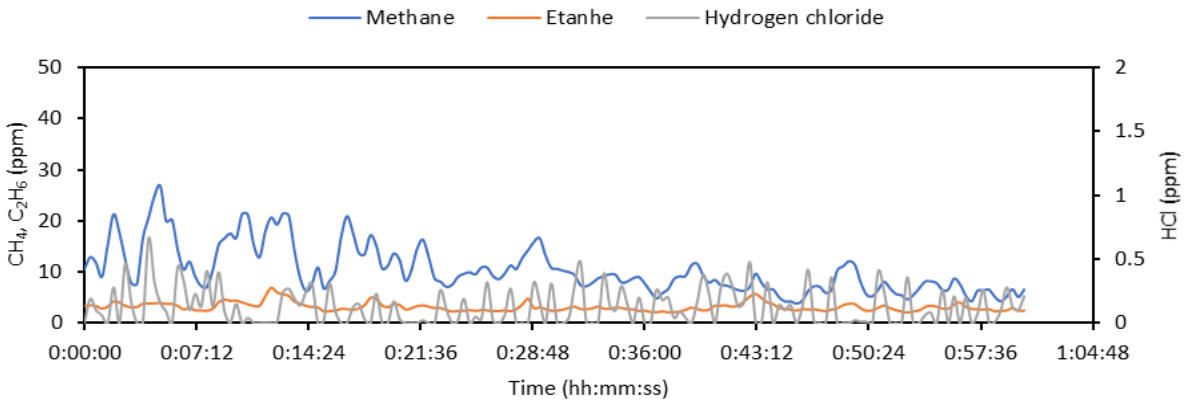

Figure 4.33. CH_4, C_2H_6, HCl concentrations in the exit flue gases along time, during combustion of BioSolids Unal pellets.

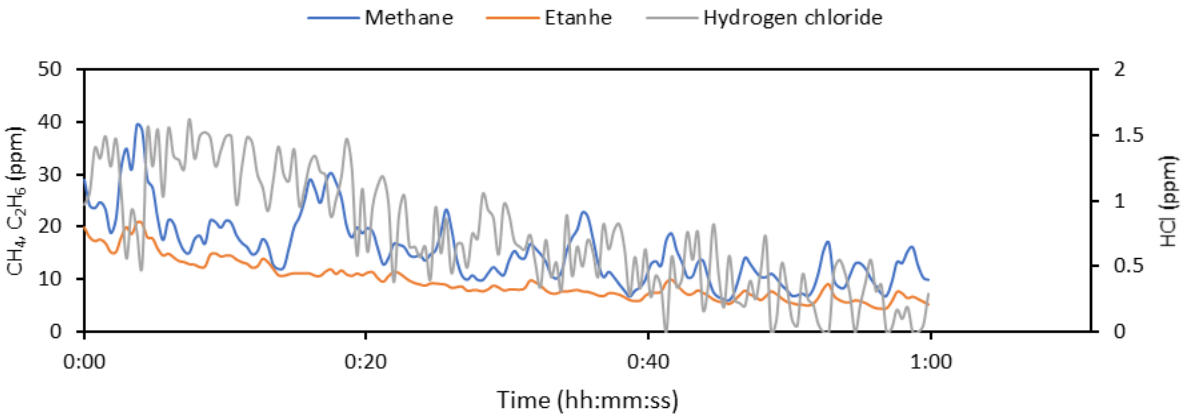

Figure 4.34. CH_4, C_2H_6, HCl concentrations in the exit flue gases along time, during combustion of BioSolids AF-2 pellets

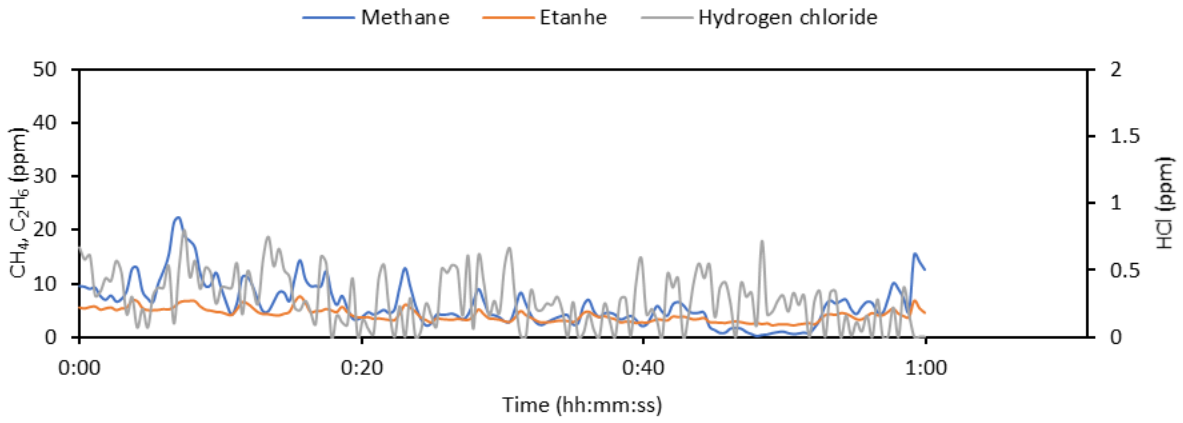

Figure 4.35. CH_4, C_2H_6, HCl concentrations in the exit flue gases along time, during combustion of Mud Unal pellets.

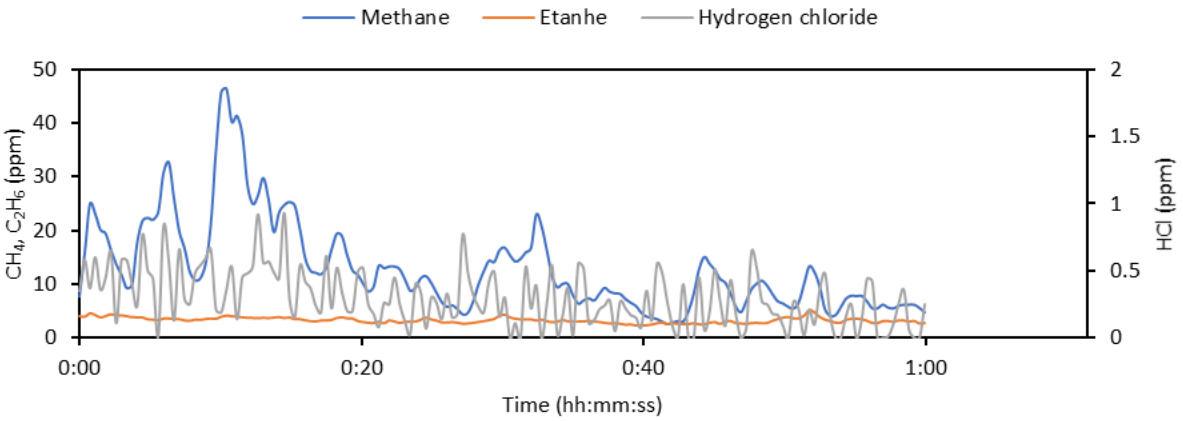

Figure 4.36. CH_4, C_2H_6, HCl concentrations in the exit flue gases along time, during combustion of BioSolids AF-8 pellets .

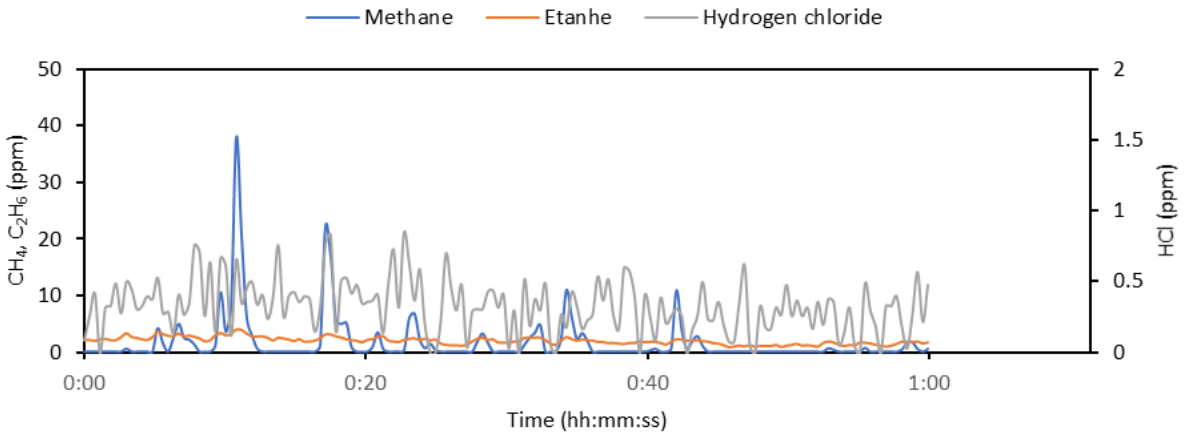

Figure 4.37. CH_4, C_2H_6, HCl concentrations in the exit flue gases along time, during combustion of torrified pellets

4.8.3 NO and SO_2 concentration in combustion flue gases

The concentration of NO presented in flue gases is closely related to the content of nitrogen in the fuel, the excess air ratio and combustion temperature. For temperatures below 1000 °C, as obtained in the experiments, the fuel NOx mechanism is the responsible for the conversion of N in the fuel into NO (see Chapter 2). The concentration of NO in flue gases along the time, for the several combustion experiments with the distinct pellet types, is shown in **Figures 4.38 to 4.45**.

In this study, torrified pellets showed the higher concentration of NO in the combustion flue gas (**Figure 4.45**), during the experiments; unfortunately, the information about the content of nitrogen in tis fuel was not available at the moment. The content of nitrogen in Pinewells pellets (0.89%wt dry basis) was higher than the content of Acacia pellets (0.78 %wt dry basis) or BioFuels Af-2 (0.70 % dry basis) (see **Table 4**), however, the NO concentration in flue gases was lower for Pinewells pellets combustion (see **Figure 4.38**). This result could be associated to influence of a lower excess air ratio (λ) during the combustion of Pinewells pellets (operated with a $\lambda = 2.75$), thus, lower than the (λ) observed during Acacia or BioSolids Af-2 combustion. This behavior is shown **Figure 4.46**, expressing the influence of the excess air on the NO concentration in flue gases. Acacia pellets also showed the higher fluctuations of NO concentration in flue gases along the time (**Figure 4.39**), and this can be related to the fuel feeding conditions but also fuel content on Nitrogen and its influence on gas composition. In fact, for the other pellets tested, the NO concentration fluctuations were of minor amplitude **(Figures 4.38**, **4.40** to **4.45**).

The presence of sulfur dioxide (SO_2) concentration in flue gases is related to the presence of sulfur in the pellets. The concentration of SO_2 in flue gases along the time, for the several combustion experiments with the distinct pellet types, is shown in **Figures 4.38 to 4.45**. The elemental composition indicated that the content of Sulfur is very low for the distinct pellet types used (see **Table 4**): below 0.1%wt (dry basis) for all the non-commercial pellets, and below 0.01 %wt (dry basis) for Pinewells pellets; there is no information available for the Sulfur content of Acacia and Torrified pellets. Consequently, it was found lower concentrations of SO_2 in the combustion flue gas of Pinewells (**Figure 4.38**).

BioSolids AF-2 pellets made from an Italian poplar variety fertilized with sewage sludge mixed with cereal straw showed the highest concentration of SO_2 in flue gases (**Figure 4.42**). For the BioSolids AF-2 and torrified pellets the SO_2 concentration was higher at the beginning of the experiment of combustion, and then decreased along time (**Figures 4.42 and 4.45**). For the other pellet types, fluctuations in SO_2 concentration along the time, did not show a decrease in SO_2 concentration along the time of the experiment. That trend for a decrease of concentration along time is a relevant issue and must subject of further research, because it can be related to some adsorption of SO_2, for example in the ash that is being accumulated in the combustion system.

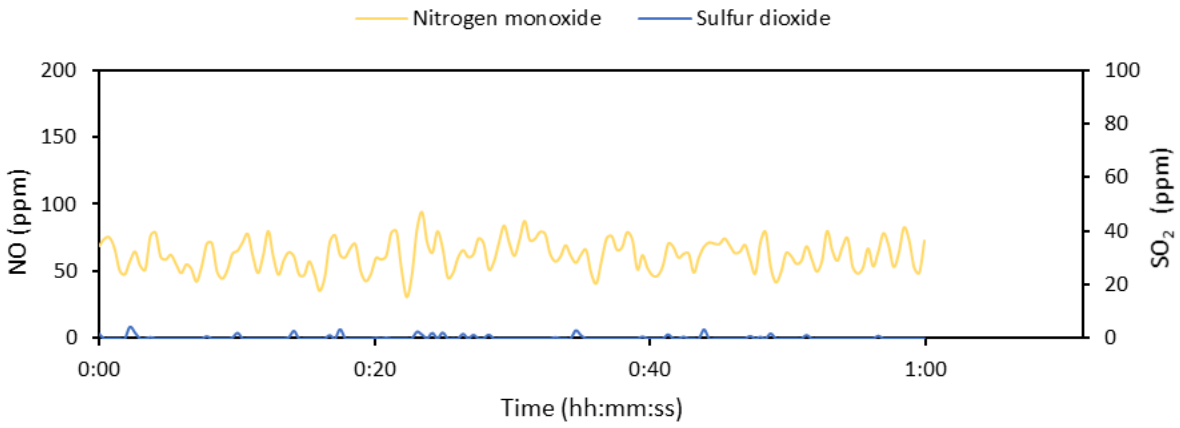

Figure 4.38. NO and SO_2 concentrations in the exit flue gases along time, during combustion experiments with Pinewells Pellets .

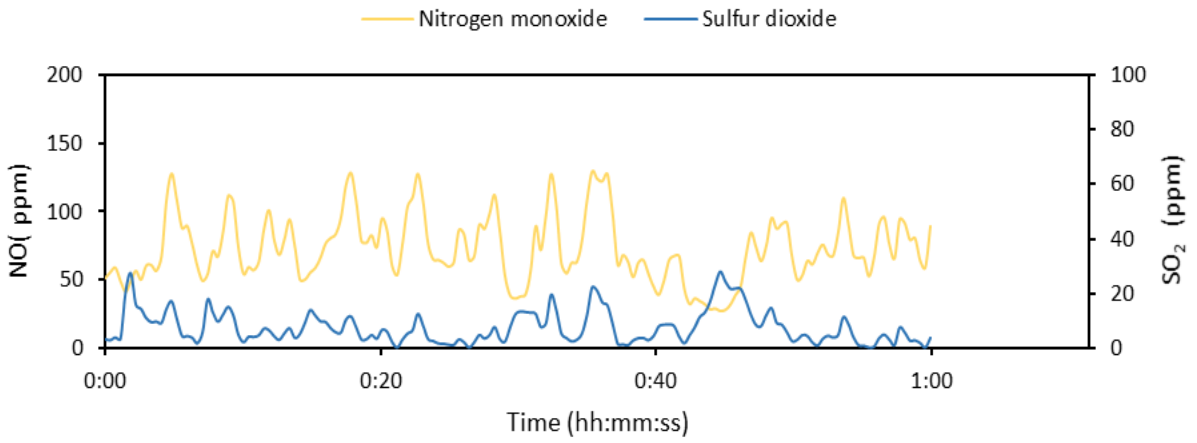

Figure 4.39. NO and SO_2 concentrations in the exit flue gases along time, during combustion experiments with Acacia pellets.

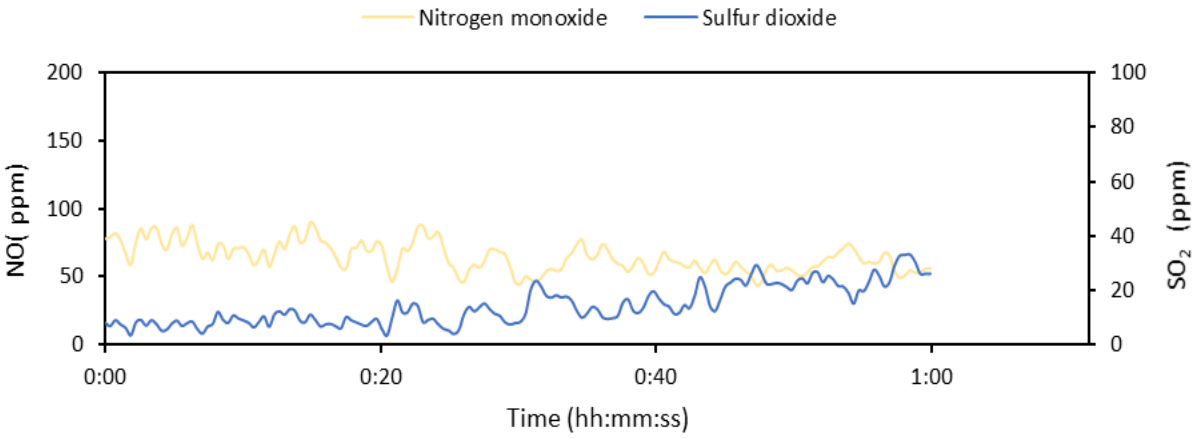

Figure 4.40. NO and SO_2 concentrations in the exit flue gases along time, during combustion experiments with Mud Af-8 pellets.

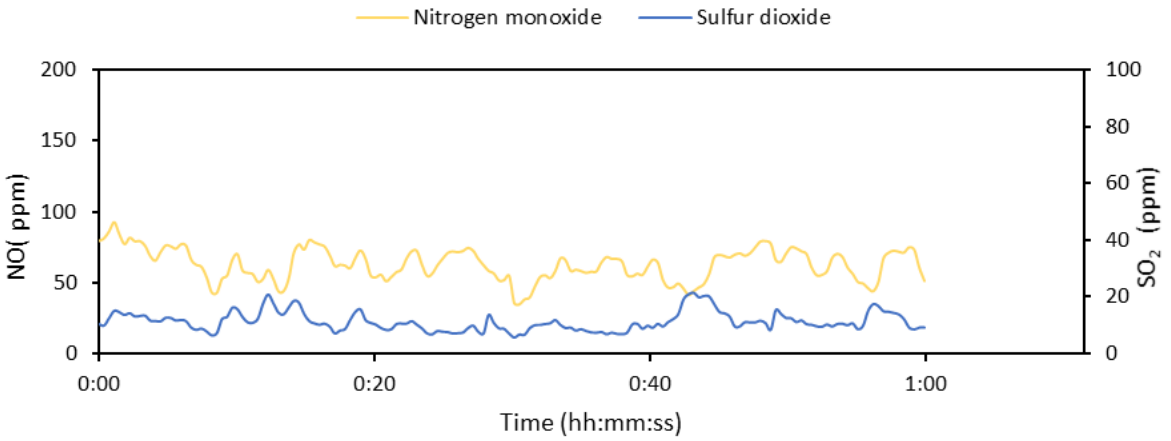

Figure 4.41. NO and SO_2 concentrations in the exit flue gases along time, during combustion experiments with BioSolids Unal.

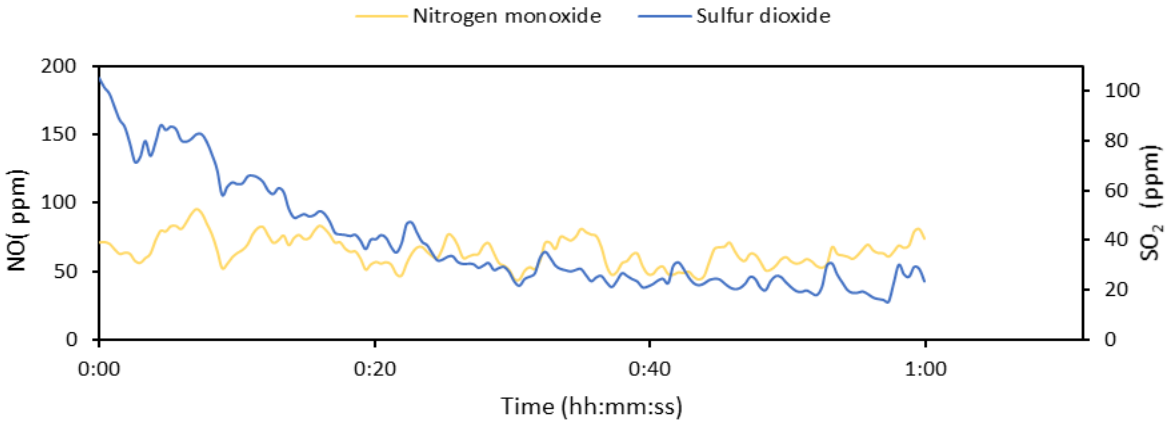

Figure 4.42. NO and SO2 concentrations in the exit flue gases along time, during combustion experiments with BioSolids AF-2 pellets.

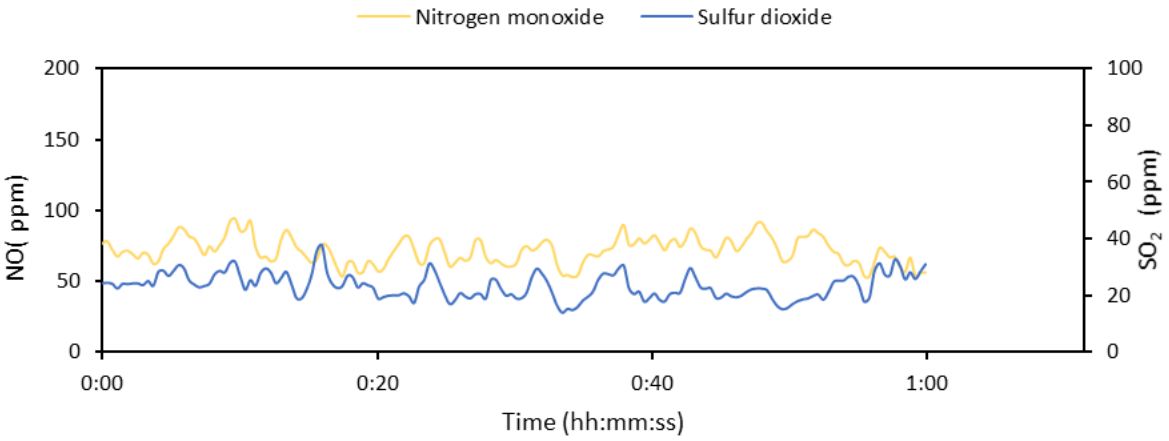

Figure 4.43. NO and SO_2 concentrations in the exit flue gases along time, during combustion experiments with Mud Unal pellets.

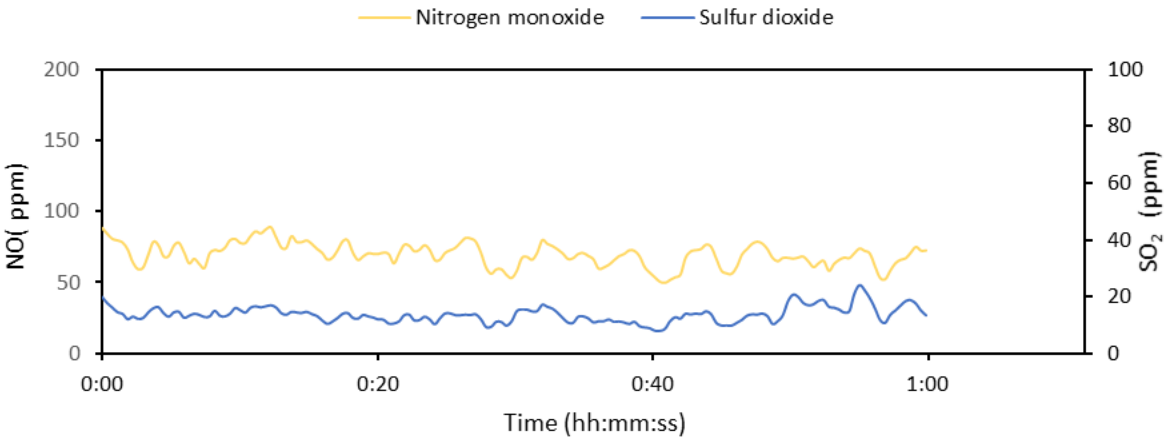

Figure 4.44. NO and SO_2 concentrations in the exit flue gases along time, during combustion experiments with BioSolids AF-8.

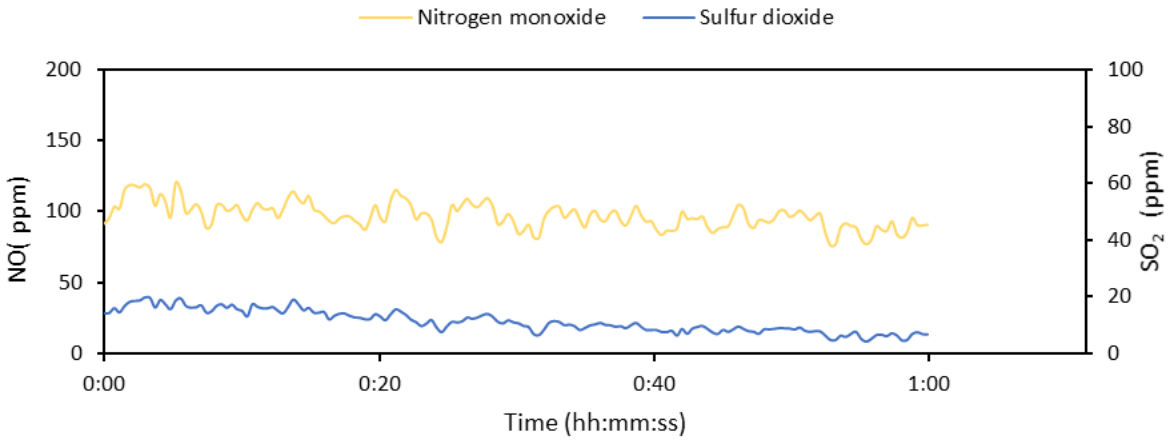

Figure 4.45. NO and SO_2 concentrations in the exit flue gases along time, during combustion experiments with torrified pellets.

4.8.3.1 NO emission as function of primary air excess ratio

It observed that the emission of NO increases with the increase of the primary excess ratio (**Figure 4.46**). Higher NO concentration in flue gases was observed in Acacia pellets 226 mgNO ·Nm^{-3} (dry gases, 11%v O_2), and during the combustion process the λ was 4.40, that is, the higher (λ) reached in all the experiments. On the other hand, during Pinewells pellets combustion the λ was 2.75 and the NO concentration in flue gases was 131 mgNO· Nm^{-3} (dry gases, 11%v O_2). The limit value of emission of NO according to the Portuguese legislation is 650 mgNO Nm^3 (dry gases, 11%v O_2) [86] as shown in **Figure 4.46**.

Typically, the NO concentration values are below 250 mg Nm^{-3} (corrected to 11%v O_2, dry gases), and show a slight increase with the excess air ratio (**Figure 4.46**), thus reflecting the influence of oxidation of Nitrogen in the fuel into NO. When compared to the Portuguese regulation 650 mg NO_2· Nm^{-3} (corrected to 11%v O_2, dry gases)[86] The calculation of the limit emission was based in the average concentration of NOx in the combustion flue gases, during the combustion period. The NO concentration in the flue gas expressed as NO_2, is below 380 mg NO_2 · Nm^{-3} (corrected to 11%v O_2, dry gases) for the combustion experiments of the several pellets. Thus, below the Portuguese regulation for combustion systems using biomass (**Figure 4.46**).

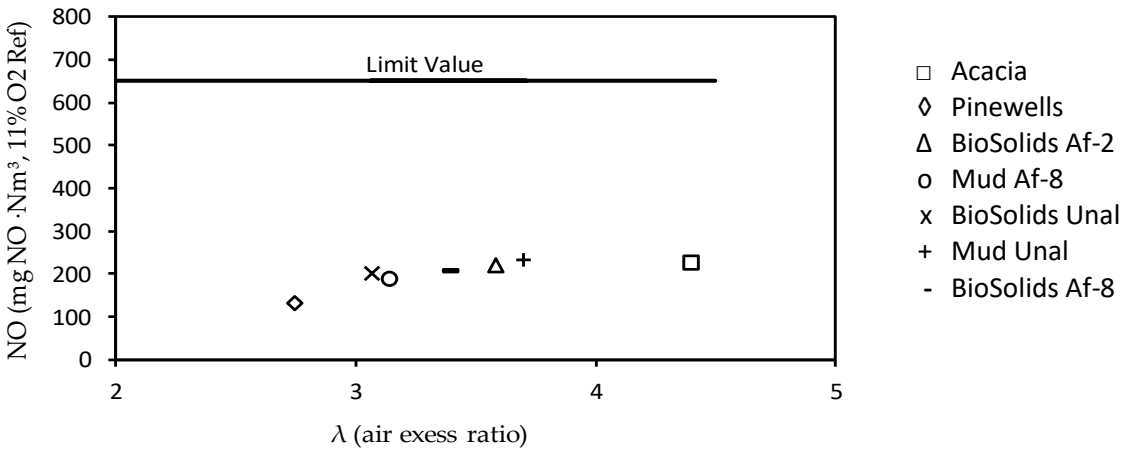

Figure 4.46. NO concentration in the flue gas expressed as NO_2 and corrected to 11%v O_2, dry gases) as a function of primary excess air ratio for different biomass pellets tested. It also show the emission limit value set by Portuguese regulation [86].

4.8.3.2 Fuel Nitrogen conversion to NO

The different fuels had different nitrogen content, and the experimental data analysis for the distinct combustion experiments shows a decreasing percentage on the fuel Nitrogen conversion to NO with increasing the Nitrogen content of the biomass fuel (**Figure 4.47**), as often referred in the literature[22][87]. A fuel Nitrogen conversion to NO below 30% was observed in this study.

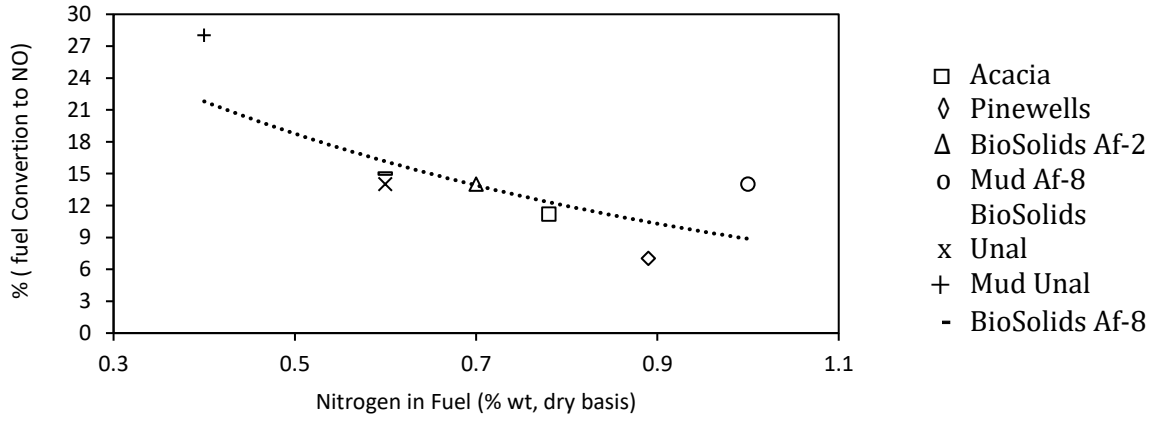

Figure 4.47. Percentage of Nitrogen content in fuel converted to NO in the combustion flue gases.

4.8.3.3 Sulphur dioxides as function of sulfur content in fuel

Sulfur dioxides have a direct relation with the amount of sulfur present in the fuel. Bio-Solids Af-2 reached the highest emission, and the lowest was reached by the Pinewells pellets, 296 and 0.30 mg SO_2 Nm^{-3} (dry gases, 11%v O_2). For this pollutant, all the pellets accomplish the limit set by Portuguese regulation. The calculation of the limit emission was

based in the average concentration of SO_2 in the combustion flue gases, during the combustion period

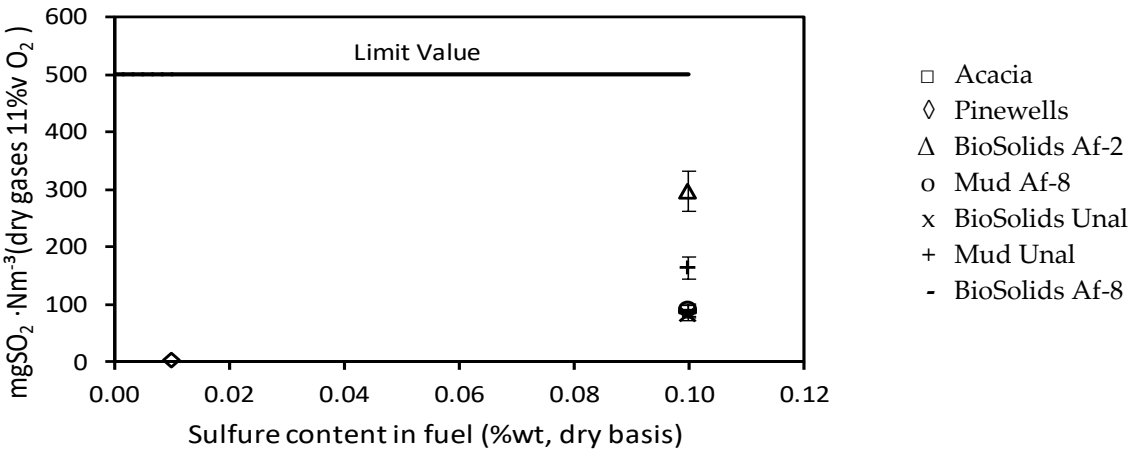

Figure 4.48. Relation between sulfur content in the biomass fuel and the SO_2 concentration in the combustion flue gas, for all the biomass pellets. It is also shown the limit of SO_2 concentration in the flue gas emission following the Portuguese legislation [86].

4.9 Pollutants concentration in flue gas during stove shut-down

It was observed that during the shut-down procedure of the stove, relatively high concentration of gases related to incomplete combustion, such as CO or C_2H_6 , CH_4, were observed in the exit flue gases, as shown for example in **Figures 4.49 to 4.50**. (**Figures 4.20** to **4.27** to CO, and **Figures 4.30** to **4.37** for CH_4 and C_2H_6). In some cases, as occurred with Unal Mud pellets the concentration of C_2H_6 was ten times higher (**Figure 4.49**) than the maximum value reached during the combustion experiment conducted (**Figure 4.35**). During normal operation combustion the mean value of C_2H_6 concentration for Unal Mud was 4.20 ppm, and increased to 34.9 ppm during the shutting down process. In the case of CO, the concentration reached during the shut-down procedure was as high as two times higher the maximum observed during the combustion experiments, as can be seen for BioSolids Af-2 pellets and BioSolids Unal pellets, in F**igures 4.50** and **4.51.**

The shut-down procedure of the stove created combustion conditions that promote the emission of such compounds, and among the reasons, the decrease in temperature in the combustion chamber to values around 400°C to 500°C is among the most probable, although some other reasoning can come into play, as the proper air/fuel mixing and availability of O_2. This issue related to shut-down procedure and related gaseous pollutant emissions must be improve in the operation of the stove.

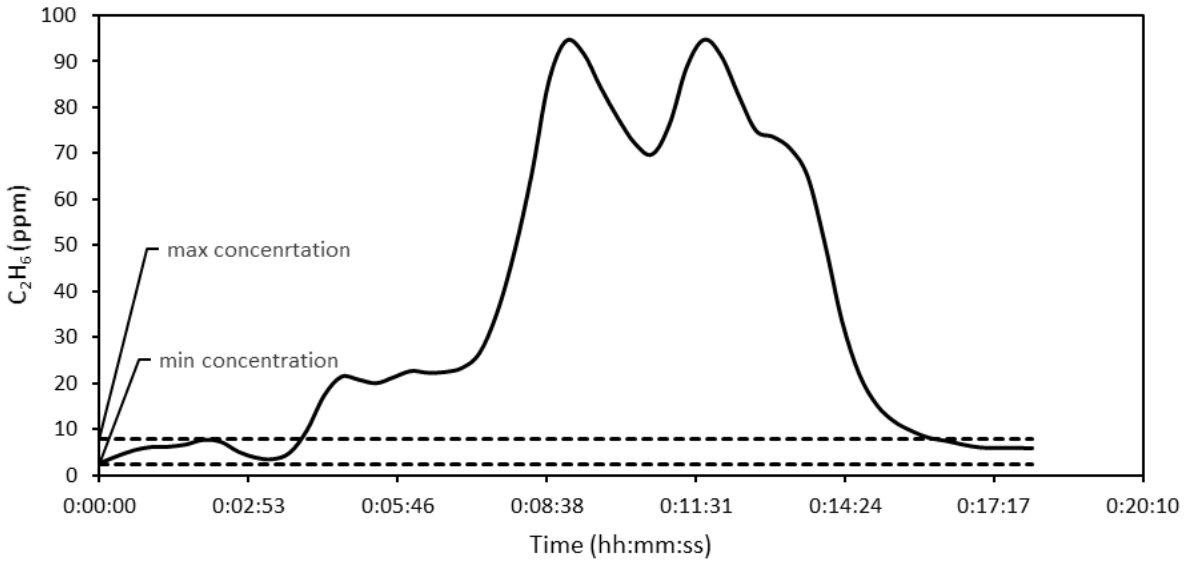

Figure 4.49. Behavior of Ethane concentration in combustion flue gases during stove shut-down, after combustion of Unal Mud pellets.

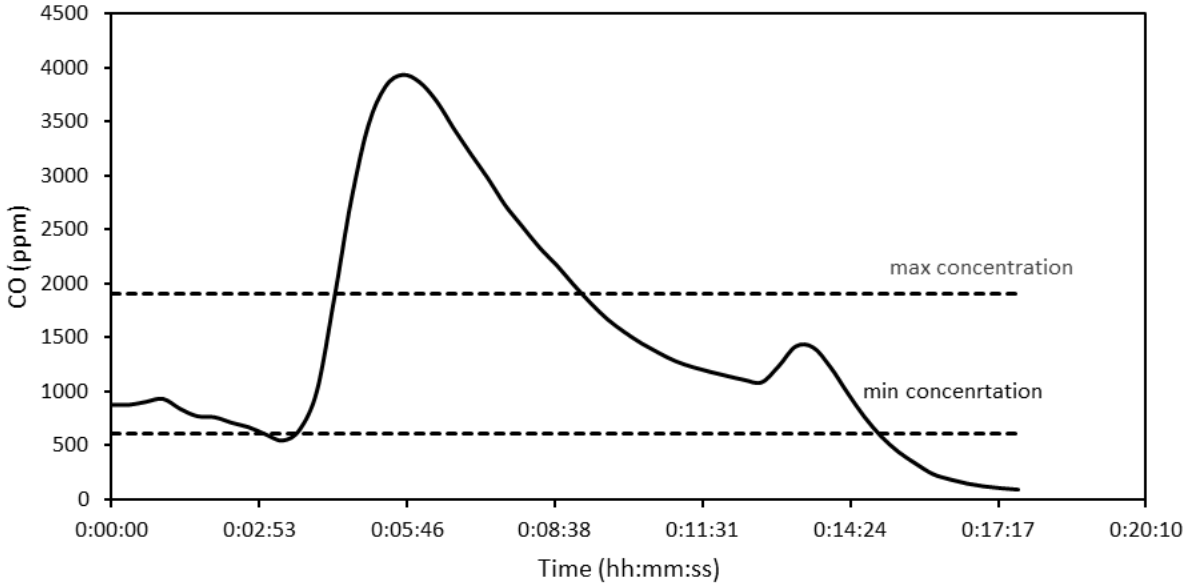

Figure 4.50 Behavior of CO concentration in combustion flue gases during stove shut-down, after combustion of BioSolids Af-2 pellets.

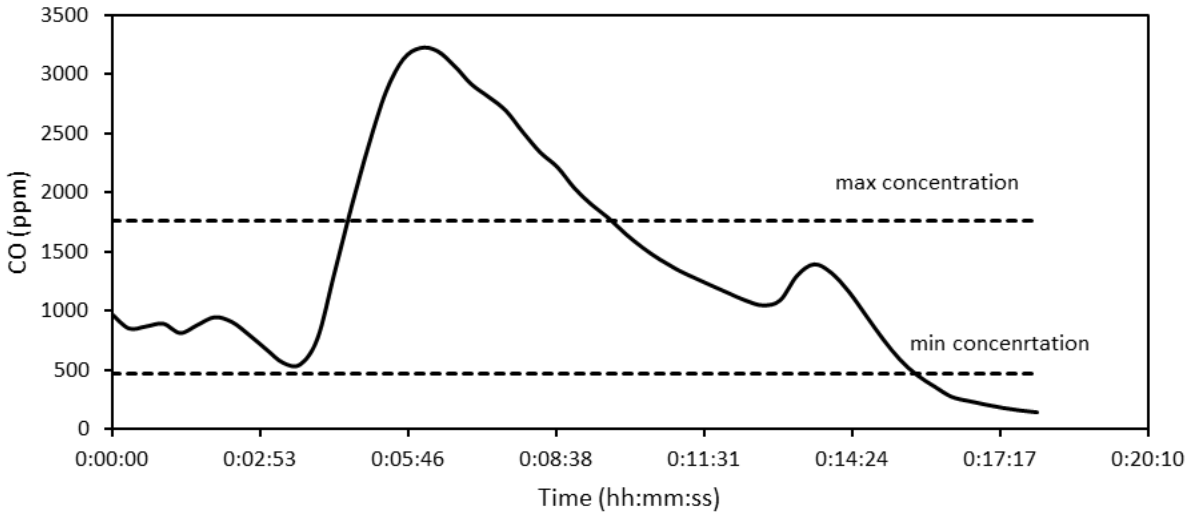

Figure 4.51 . Behavior of CO concentration in combustion flue gas during stove shut-down, after combustion of BioSolids Unal pellets.

4.10 Emission factors.

The emission factors (EF), expressed as $mg_{pollutant}/kg_{fuel\ db\ consumed}$, were calculated considering the mean concentration of the gaseous compound in the combustion flue gas, and the combustion gas flow rate in the chimney. Equation 8 adapted from Venturini et al [88] was used to calculate the emissions factors (EF) in [$mg \cdot kg^{-1}{}_{fuel\ dry\ basis}$].

$$EF_y = \frac{\sum \overline{Cy} \times \overline{\dot{V}_{fg}}}{\dot{m}_f} \quad \text{(Eq.8)}$$

The distinct physical and chemical characteristics influenced the reactions during combustion, as a consequence flue gas composition observed for every biomass pellets was different. The EF obtained for all pellets tested was always higher during the combustion of the noncommercial, non-certificated pellets.

It can be observed in that the Carbon Monoxide's EF from Pinewells and torrified pellets were the lowest (**Figure 4.52)**. Previous studies [89] showed carbon monoxide EF 's values ranged between 1636 $mgCO \cdot kg^{-1}$ to 13320 mg $CO \cdot kg^{-1}{}_{(db\ fuel).}$ The CO EF's obtained in this study was in the range between 3582 to 21597 $mgCO \cdot kg^{-1}{}_{(db\ fuel).}$ Torrified pellets achieved best EF 580 $mgCO \cdot kg^{-1}{}_{(db\ fuel).}$ Pellets made of a wood mixture with straw as the Mud and the BioFuels were the ones that showed the higher emission factors. A typical characteristic between MUD and BioFuels is the high content of ash.

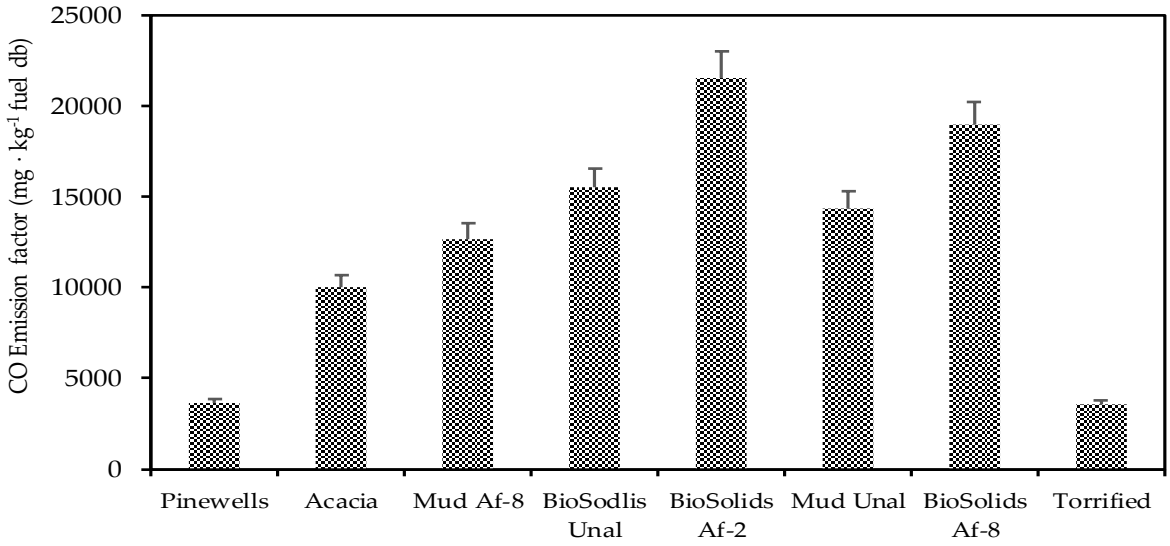

Figure 4.52. Carbon monoxide (CO) emission factors ($mgCO \cdot kg^{-1}$ biomass consumed db) from pellets combustion in the stove in operated in manual mode (parameters conditions: Fire 3 and Fan 3).

Focusing on CH_4 and C_2H_6 emission factors, **Figures 4.53, 4.54** shows the emission factors of calculated for all the combustion experiments conducted. The highest methane and ethane emission factors values occurred in the combustion of BioSolids AF-2 pellets. The CH_4 and C_2H_6 emission factors were higher when combustion conditions such temperature in the combustion chamber was lower. Lesser EF were obtained for the combustion of pinewells, torrified and Mud Af-8 pellets. It was observed common combustion parameters during the experiments with Pinewells, and Mud Af-8, such as lowest primary excess and mean combustion temperatures in location T1 over 650 °C. Torrified pellets show similar EF's as Pinewells pellets (**Figure 4.53** e **4.54**). Due to the lack of information about the chemical composition of torrefied pellets, it was not possible to determinate primary excess air ratio in that case, but the measured mean temperatures in the combustion chamber reached 850°C. High emission factors of C_2H_6 CH_4 are a clear indication of poor combustion conditions.

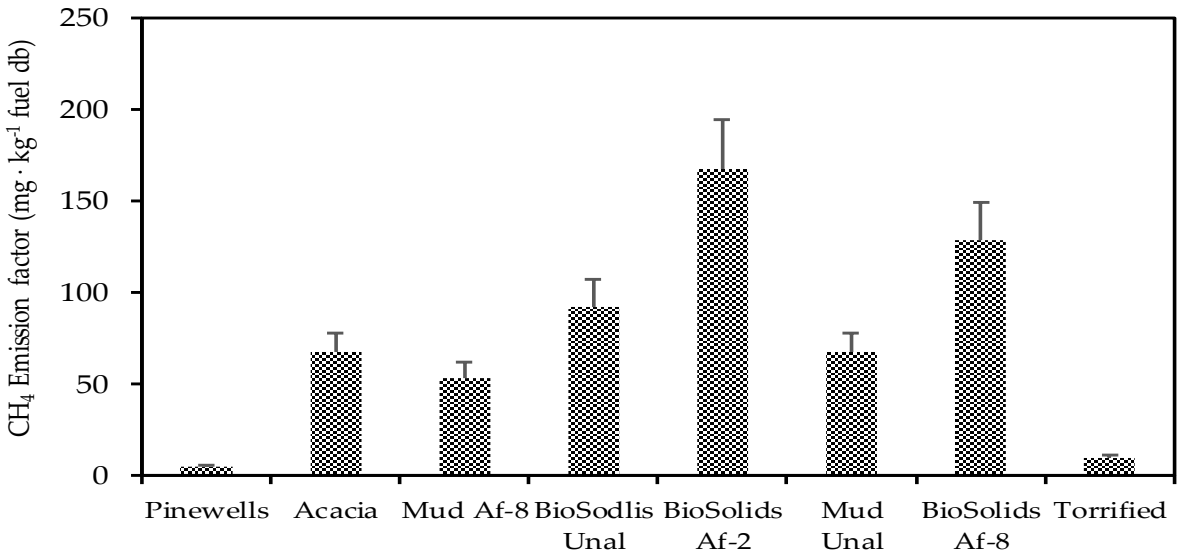

Figure 4.53. Methane (CH_4) emission factors (mg $CH_4 \cdot kg^{-1}$ biomass consumed db) from pellets combustion in the stove in operated in manual mode (parameters conditions: Fire 3 and Fan 3).

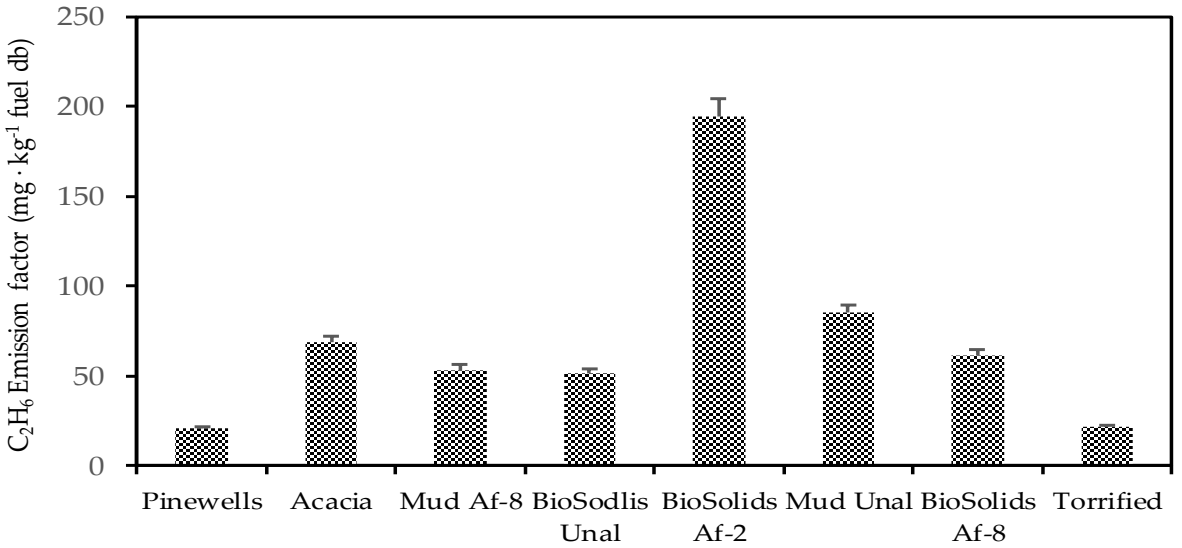

Figure 4.54. Ethane (C_2H_6) emission factors (mg$C_2H_6 \cdot kg^{-1}$ biomass consumed db) from pellets combustion in the stove in operated in manual mode (parameters conditions: Fire 3 and Fan 3).

Differences between the SO_2 EF 's were mainly due to the different content of Sulfur in the biomass fuel. The elementary analysis showed content values of sulfur very low and

similar for all the pellets tested, however, the EF calculated showed values ranging from 2 to 1707 $mgSO_2 \cdot kg^{-1}$ (db fuel) (**Figure 4.56**). BioSolids AF-2 had the highest SO_2 EF, and it could be related to the fact that more of the SO_2 generated was released as gas into the flue gas, and for the other pellets SO_2 can be adsorbed and retained in the ashes; this subject must be object of further studies to be made clearer.

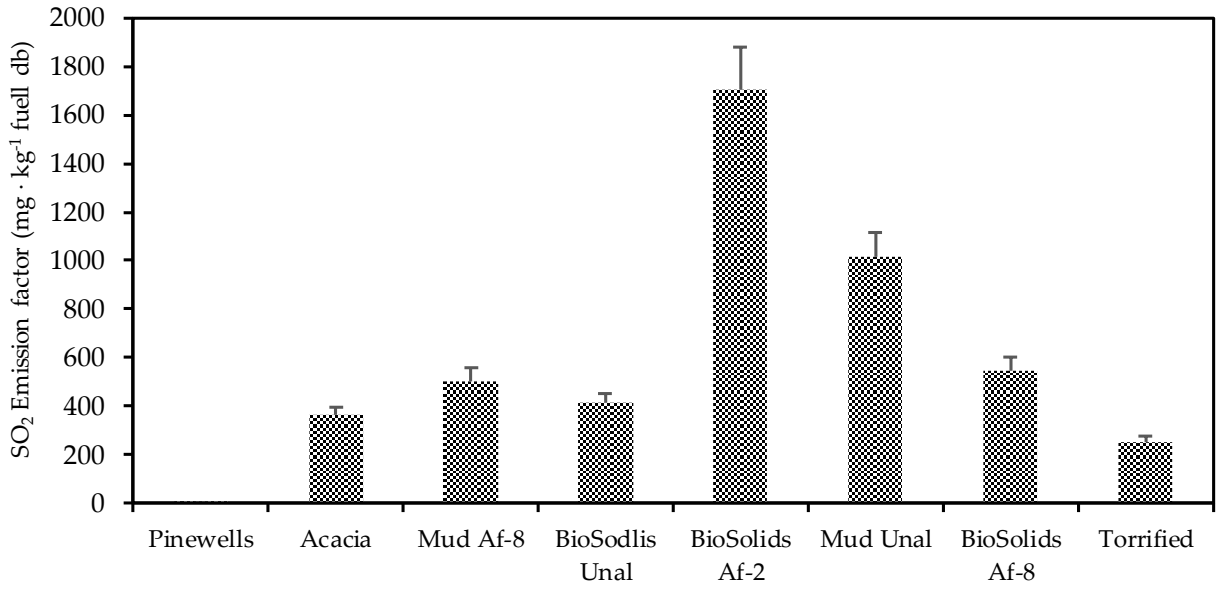

Figure 4.55. Sulfur dioxide (SO_2) emission factors ($mgSO_2 \cdot kg^{-1}$ biomass consumed db) from pellets combustion in the stove in operated in manual mode.

5. Conclusions.

This study aimed at characterizing the operating conditions of eight types of biomass pellets by using a stove for residential heating. The experiments showed the influence of physical and chemical characteristics of pellets in the operating combustion conditions, such as the flue gas temperature, primary air flow rate, primary excess of air ratio (λ), fuel consumption rate, among others. Different operating conditions in the stove changed the composition of the combustion flue gas.

The behavior of temperature in the combustion chamber was influenced by two factors. On the one hand, the physical and chemical characteristics of the biomass pellets such us, length, poor consistency, ash and moisture content. An on the other the feeding system of the stove. Moisture content in pellets reduces the possible maximum combustion temperature. The poor consistency of pellet turned them in dust and particles, making even harder to the feeding system, to provide continuous and uniform feeding of pellets into the combustion chamber. Thus, generating fluctuation in the temperatures profiles. Pellets with ash content superior to 2.5 %wt ash (dry basis), such as Mud Af-8, BioSolids Unal, Biosolids Af-2 caused ash accumulation in the grate of the stove and disturbed the proper circulation of the primary air flow rate through the fuel bed. As a result, the combustion of these pellets presented a higher concentration of some pollutants in flue gas compared to. In addition, after 60 minutes of continuous function, the ash produced by these pellets blocked the exit of the feeding port interrupting the operation.

Higher fuel consumption was associated with elevated temperatures in the combustion chamber and with an operation of the stove with a lower primary air flow ratio. Commercial torrified pellets reached the maximum fuel consumption 1.40 $kg \cdot h^{-1}$, producing the higher temperatures observed in the combustion chamber. Whereas, combustion of Mud Unal pellets registered the lowest fuel consumption 0.75 $kg \cdot h^{-1}$.In general, the behavior of the mass of the fuel followed decreasing linear tendency. Small variations in this linear tendency can be associated with some fluctuations in the stove's feeding system because to the heterogeneity of the physical properties of the pellets.

The excess of air is an essential operating characteristic of domestic combustion technology because it reduces CO emission. To achieve this reduction, domestic systems usually operate with high values of excess of air (e.g., $\lambda > 2$). For this study, the excess of air was in the range of λ=2.75 and λ=4.4. The CO emission and primary excess air ratio showed a pattern; the increase of primary air excess ratio (λ) increases CO concentration of in combustion flue gases.

The current shut-down procedure created combustion conditions that promote the emission of CO, CH_4, C_2H_6. For example, as occurred with Unal Mud pellets the concentration of C_2H_6 was ten times higher than the maximum value reached during the combustion experiment conducted. Unal Mud pellets showed a maximum concentration of 7.64 ppm during normal operation of combustion and increased to 94.9 ppm during the shutting down process. Therefore, findings show that the operation of the stove can benefit from improving the shut-down procedure. It was observed that factors such as the low temperature and high

primary excess ratio, which promoted the CO formations, also influenced the formation of CH_4 and C_2H_6.

To conclude, the experiments conducted with commercial, and certified pellets, reached higher temperatures, had better steady operation and lower pollutant emission factors. Whereas the use of non-commercial and non-certified pellets, with a higher content of ash and moisture, maximized the emission of pollutants. The emission factors obtained for all pellets tested was always higher during the combustion of the non-commercial, non-certified pellets. Hence, it is highly recommended that quality control measures are applied before using non-commercial pellets in stoves for domestic use.

Future Research

In addition to the experimental tests performed, further research can be focused on conducting an economic and energetic analysis of the use of torrified pellets. Moreover, this study can be complemented with a combustion characterization and comparison of torrified and white pellets.

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